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[*Authors alone are responsible for all opinions expressed in their Communications.*]

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XXI.

THE CONCENTRATION AND PURIFICATION OF ALCOHOLIC FERMENTATION LIQUORS.

Part I.—The Distillation in Steam of Certain Alcohols.

By JOSEPH REILLY, M.A., D.Sc., M.R.I.A., F.I.C.,

AND

WILFRED J. HICKINBOTTOM, M.Sc., A.I.C.

[Read January 25. Published August 19, 1921.]

The monohydric aliphatic alcohols may be considered as being formed from water by the substitution of an alkyl group for one of the hydrogen atoms. They may also be regarded as hydroxyl derivatives of the saturated paraffins. From the former standpoint the water-alcohol mixtures would be of the type of *closely-related* liquids. According to the latter view, while paraffin-alcohol mixtures would conform to this type, aqueous alcohol compositions would more readily be classified among mixtures of *not-closely-related* liquids. From both these aspects the subject has been studied by Young[1] in his investigation of the relationship of the alcohols to water on the one hand, and to the paraffins on the other. In general the influence of the alkyl group rapidly increases as we pass from methyl alcohol up the series. Furthermore, from the study of the vapour pressures of various mixtures of alcohols and water,[2] it has been shown that the relation between the molecular percentage of methyl alcohol in the mixture and the corresponding vapour pressure may be represented by an approximately straight line. Comparisons were made by Young from Konowalow's data at such a temperature that the vapour pressure of the pure alcohol in each case was 400 mm. With the substitution of ethyl alcohol for methyl alcohol in the above comparison, the relation is represented by a decided curve. In the case of the higher alcohols the curve becomes still more pronounced.

It would, therefore, appear that methyl alcohol water mixtures come into the group of *closely-related* miscible liquids, while ethyl alcohol and water mixture is better classified with the miscible *not-closely-related* mixtures.

[1] Trans. Chem. Soc., 1902, 81, 707. [2] Konowalow, Weid. Ann., 1881, 14, 34.

In the case of some of the higher alcohols obtained by fermentation, such as *iso*-butyl and *iso*-amyl alcohols, only partly miscible mixtures are obtained with water, and it is more convenient to classify these as *partly miscible, not-closely-related,* mixtures.

Investigations on the distillation of homogeneous mixtures were first carried out by Gay-Lussac,[1] and subsequently by Magnus,[2] Regnault,[3] Berthelot,[4] and others. This early work was correlated by Duclaux, who used it as the basis of experimental work, which led to the establishment of a relation between the composition of the liquid to be distilled and the distillate. He states[5] that ethyl alcohol distils from aqueous solution more slowly than amyl or *iso*-butyl alcohols.

One of the processes adopted for the purification of ethyl alcohol on a technical scale consists in the distillation of a dilute aqueous solution of the "raw spirit."[6] Under these conditions the fusel oil, various esters, and other impurities are removed more readily than the ethyl alcohol, and in consequence are collected in the first running. The dilute solution of ethyl alcohol collected in the subsequent fractions is then concentrated. The process depends on the fact that the higher alcohols distil from dilute aqueous solution at a more rapid rate than does ethyl alcohol.

Duclaux studied the rate of distillation of the alcohols with diminishing volume. He took a certain volume of aqueous alcohol and distilled it, collecting the distillate in several fractions. By expressing the volume of the alcohol as a percentage of the total amount distilled he obtained a series of constants for each alcohol which served to identify the particular alcohol.

As the original method outlined by Duclaux does not give concordant results with different workers, a large amount of experimental work has been carried out to remedy the discrepancies observed. It has been shown that the loss of heat from the exposed flask and still-head, by convection currents and radiation, leads to irregular results. Various arrangements for preventing this have been used. It is not, however, advisable to keep the exposed flask at a higher temperature than that of the vapour, otherwise errors will be introduced by splashing and by complete evaporation of drops of solution. The best conditions for preventing condensation are obtained by surrounding the flask with a steam jacket.

The original formula of Duclaux may be expressed thus:—

$$\frac{da}{db} = c\frac{a}{a+b},$$

[1] Ann. de Physique et de Chimie, 1815.
[2] Ann. de Physique et de Chimie, 1836.
[3] Phil. Mag., 1855, 9, 4.
[4] Comptes rendus, 1863, 57, 430, 985.
[5] Ann. inst. Past., 1895, ix, 575.
[6] E. Guillaume, E.P., 5194, 1902.

where a and b represent the percentage by volume of alcohol and water respectively in the original liquid, and da and db the percentage of alcohol and water respectively in the vapour. The above relation may be represented by a hyperbola. The value for c with dilute solutions of various alcohols varies from 10·9 for methyl alcohol to 50 for amyl alcohol and 61 for capryl alcohol. The coefficient c increases with the molecular weight of the alcohol. The relation established by Duclaux has been verified for ethyl alcohol and several of the higher alcohols up to the point corresponding with mixtures of constant boiling point.

Sorel,[1] pursuing the problem from the point of view of its industrial application, carried out distillations on quantities of four litres of liquid. He took precautions to prevent loss of heat by radiation, and probably it is due to this fact that his results differ somewhat from those of Duclaux. Against the volume distilled, Sorel plotted the composition of the remaining liquid, and obtained a curve representing the rate of elimination of alcohol from the aqueous solution.

The distillation of dilute solutions can be represented approximately by the expression

$$Va = (V - dV)(a - da) + U dV,$$

or

$$U = a + V\frac{da}{dV},$$

where

V = volume remaining in the flask at any moment;
a = concentration of alcohol in the solution;
U = concentration of alcohol in the distillate.

Mariller[2] uses the coefficient K, which is given by

$$K = \frac{\text{percentage of alcohol in vapour}}{\text{percentage of alcohol in liquid}},$$

or, in the Duclaux notation (see above), $= \dfrac{da}{a} = \dfrac{100\,c}{ac + 100}$.

It is found that the coefficient K, which may be called the coefficient of solution, or better, the coefficient of enrichment, is dependent on the alcoholic concentration. It is found that on Sorel's formula, $K = 9{\cdot}9$ for ethyl alcohol of 1 per cent. concentration, and this value gradually diminishes as a increases. With $a = 20$ $K = 3{\cdot}31$, with $a = 50$ $K = 1{\cdot}5$, and, as a increases, the value of K gradually falls from 1·5 to 1. Thus for $a = 96$, $K = 1{\cdot}002$, and for $a = 97$, K is recorded as 1·001, and for $a = 100$, K is given as 1. The last two values are apparently incorrect, for it would be expected that K should

[1] Comptes rendus, 1892, 116, 693.

[2] "La Distillation Fractionnée," 1917, p. 23: cf. also "Le Bulletin de l'Assoc. des Chimistes," 1911, p. 473, &c.; and "La Distillation," p. 147.

equal unity for a concentration of 95·47, while for higher concentrations K should fall to less than unity.

From the industrial point of view it is necessary to know, during the purification of the alcohols, the rate at which the impurities are removed with respect to the ethyl alcohol.

If s be the weight of impurities in part of kilogram per kilogram of the original liquid, and S the weight of impurities in 1 kilogram of the mixed vapour, Sorel gives the following relationship:

$$S = K_1 s + K_2 s^2 + K_3 s^3 \ldots;$$

or $$S = K_1 s \text{ approximately.}$$

The distillation of mixtures of ethyl alcohol, higher alcohols, various esters, aldehydes, &c., and water over a wide range of alcohol concentration has been made by Barbet.[1]

The ratio

$$\frac{\text{Percentage of "impurities" in ethyl alcohol in distillate}}{\text{Percentage of "impurities" in ethyl alcohol in liquid}}$$

he designated by K', and it is known as the coefficient of purification. If

$$k' = \frac{\text{percentage by wt. of impurities (i.e. amyl alcohol, \&c.) in vapour}}{\text{percentage by wt. of impurities (amyl alcohol, \&c.) in liquid}},$$

then $K' = \dfrac{k'}{K}$.

The value for K' is dependent largely on the alcoholic concentration of the solution. It indicates how far distillation will remove this impurity from the alcohol, apart altogether from the quantity of water present. This "coefficient of purification" is of more technical value than the coefficient of enrichment.

Using a similar notation to that employed by the authors in the study of the fatty acids,[2] the K of Sorel may be expressed in a slightly different form where

a = initial amount of alcohol in flask;

x = amount of alcohol in distillate after volume v has distilled

V = constant volume of liquid in flask.

If ρ = density of water vapour in the flask, and σ = weight of water per unit volume of distillate, we have

$$K = \frac{\text{concentration in vapour phase} \times \sigma}{\text{concentration in liquid phase} \times \rho}$$

$$= k\frac{\sigma}{\rho}, \qquad \lambda = k\frac{\sigma}{\rho V},$$

$$= \lambda V \text{ or } A \times V \times 2{\cdot}3026, \text{ where } A = \frac{1}{v}\log\frac{a}{a-x}.$$

[1] "La Rectification et les colonnes Rectificatrices," 1895, p. 46.

[2] Sci. Proc. R.D.S., 1919, xv, 519.

Using this expression, the values of the coefficient of Sorel corresponding to those of $\frac{1}{v} \log \frac{a}{a-x}$ have been calculated for solutions of alcohol of different concentrations (see later).

The distillation coefficients of the alcohol change with the concentration. Only a few results are given showing the change of constant with concentration; but these confirm the results of Sorel and Gröning. With dilute solutions of the acids the alteration in the rate of distillation from an aqueous solution can be satisfactorily explained by taking into consideration the degree of ionic dissociation. For the alcohols this explanation cannot apply. The alcoholic solutions are generally more concentrated than those employed in investigations on the acids, consequently the temperature of the aqueous solution may be a determining factor. Another possible disturbing effect may be due to the association of the alcohol. Murray[1] has adduced evidence that the molecules which are usually associated in the liquid state are generally not associated in aqueous solution. If there still exist in the solution some associated molecules at the concentration employed, there will be a continual change in the state of molecular aggregation as the distillation proceeds. Under these conditions Nernst's law of distribution will not hold, consequently the distillation coefficient may vary.

As a general rule, the application of Nernst's law to the distillation of solutions furnishes us with a method of determining whether a substance behaves normally in solution. Water has been used in all the investigations, chiefly on account of the convenience. Other solvents, such as alcohols, amines, or any other substance which is sufficiently volatile to carry over the solute in the vapour may also be employed.

In the experiments on the distillation of alcohol-water mixtures referred to, the distillations have all been carried out at varying volumes. In the present paper, however, the authors have investigated the distillation coefficients of methyl, ethyl, *n*-propyl, *n*-butyl, *iso*-butyl, *sec*-butyl, and *iso*-amyl alcohols obtained by distilling constant volumes of dilute aqueous solutions of the alcohols.

Method of Distillation employed.

The distillation of dilute solutions of the lower alcohols was carried out at constant volume in a similar type of apparatus to that employed by us for the distillation of the fatty acids. 200 c.c. of a dilute aqueous solution of an alcohol were distilled, and the volume of liquid in the flask kept constant

[1] Amer. Chem. Jour., 1903, **30**, 193.

throughout the experiment by adding water at the same rate as the solution distilled over. The distillate was collected in approximately equal fractions of 5 or 10 grams. The distillation was carried out at the approximate rate of 60 c.c. per hour. It was found convenient to employ a set of receivers, which were weighed and provided with stoppers. Before a distillation the cylinders were thoroughly cleaned with a solution of chromic acid in concentrated sulphuric acid, and finally washed several times with distilled water. They were then drained by inverting them in a clean beaker. Immediately after a fraction had been collected, the cylinder was removed and stoppered until a determination of the alcohol could be carried out.

The Alcohols investigated.

The alcohols were carefully purified by repeated fractionation, and usually a fraction boiling within 0·2° was reserved.

Methyl Alcohol was obtained from the purest commercial methyl alcohol free from acetone and ethyl alcohol. A sample was collected after two fractionations from an eight-section "Young evaporator fractionating column." It boiled at 64·4–64·6° at 759 mm. pressure. Density = 0·7965 at 15·5/15·5°.

Ethyl Alcohol was purified by refluxing over solid potassium hydroxide, then over freshly ignited quicklime, and finally over calcium. On fractionation the alcohol was obtained of density 0·7937 at 15·5/15·5° and boiling at 78·3–78·4°/761 mm.

n-Propyl Alcohol was obtained from fusel oil by repeated fractionation. The distillate collected between 97 and 97·5° 760 mm. was reserved for the determination of the distillation constant. Although a considerable quantity of fusel oil was fractionated, using both a Young column and a long fractionating column with Lessing rings (copper), only a very small proportion of alcohol with a boiling point 97·2/763 mm. and density 0·8078 at 15·5/15·5° was obtained. The purity of this alcohol is probably not of the same order as in the case of the other alcohols examined.

n-Butyl Alcohol was obtained by fermentation, and after drying it thoroughly over quicklime and potassium carbonate, it was fractionally distilled several times—until the density showed no alteration on further treatment. Boiling point = 117·6°/760 mm.; density = 0·8137 at 15·5/15·5°.

sec-Butyl Alcohol was obtained from *n*-butyl alcohol. The normal alcohol was converted into β-butylene, which was absorbed by moderately concentrated sulphuric acid under pressure yielding *sec*-butyl hydrogen sulphate. The alcohol was obtained by hydrolysis, and was purified by drying and

repeated distillation. Boiling point, 99·4–99·5°/755 mm. and density 0·8114 at 15·5/15°.

iso-Butyl and iso-Amyl Alcohols were purified by repeated fractionation of the purest commercial alcohols obtainable. The *iso*-butyl alcohol had a boiling point of 107·8°/758 mm., and density 0·8063 at 15·5/15·5°, while the *iso*-amyl alcohol boiled at 130·5°–131·5°/757 mm., and had a density of 0·8158 at 15·5/15·5°.

Estimation of the Alcohols in the distillate.

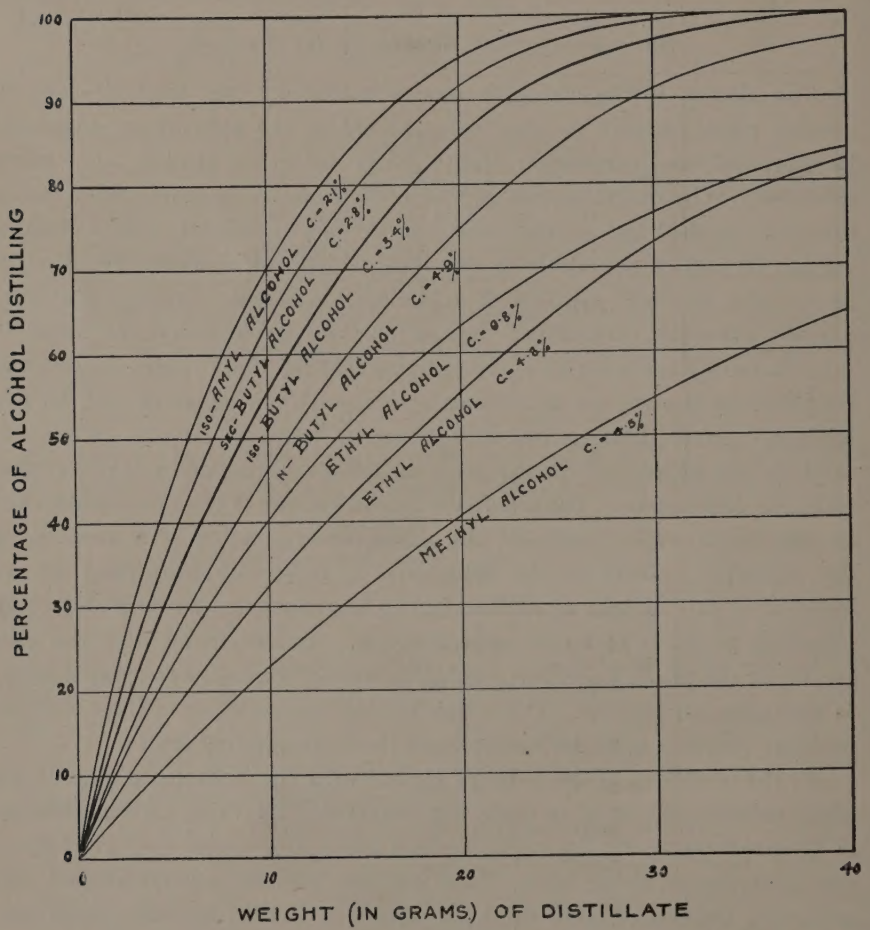
The alcohol in the distillate was estimated by two methods—(*a*) by density, using Perkin's modified arrangement of the pyknometer suggested by Sprengel, and calculation from density tables of alcohol and water mixtures; (*b*) by oxidation, the method of Benedict being employed. In the oxidation method the analysis was carried out as follows. To a known volume (2 c.c.) of the alcoholic distillate, which had been suitably diluted, 20 c.c. of a standard solution of potassium dichromate in sulphuric acid solution was added cautiously. It was found advisable to cool the alcoholic solution during the addition of the chromic acid solution. The oxidation was completed by heating the mixture on a vigorously boiling water bath for five minutes. After diluting the acid-oxidation mixture, ferrous ammonium sulphate was added, and the excess of the latter determined by titration with standard dichromate. The difference in the volume of the dichromate used for the blank experiment and the actual determination is a measure of the alcoholic content of the solution. A factor for converting c.c. of dichromate into weight of alcohol had to be determined for each alcohol by preparing solutions of known concentration. It was found that the constants for the primary alcohols, excluding methyl alcohol, were proportioned to the molecular weights. This indicates that the oxidation of these alcohols probably proceeds with the formation of the corresponding acid.

In the treatment of the *n*-butyl alcohol with the dichromate reagent, an odour resembling that of an ester was observed. By using an insufficiency of the oxidising agent, the odour became more pronounced, due evidently to greater formation of the ester. This reaction between *n*-butyl alcohol and potassium dichromate and sulphuric acid was studied in some detail (see later). When the alcohols were only partially soluble in water, a determination of the volumes of each layer was used as an alternative method in determining the alcoholic content of the distillate. In the case of *n*-butyl alcohol, by the use of the *n*-butyl alcohol-water density curve already prepared

by one of the authors,[1] it was possible to estimate the alcohol after definite dilution.

Rates of Distillation

The experimental results are given in Tables I and II, while the percentage of alcohol distilling in each fraction is shown graphically in the figure.



[1] Sci. Proc. R.D.S., 1919, xv, 43, 598.

TABLE I.

Alcohol. Initial volume = 200 c.c.	Concentration in grams of Solute per 100 c.c.	Weight of Distillate.	Percentage of Alcohol collected in Distillate.	$\frac{1}{v}\log\frac{a}{a-x}$
Methyl,	4·5	10·14	23·2	0·0113
	,,	20·01	40·5	0·0113
	,,	29·97	55·1	0·0116
	,,	50·53	72·8	0·0112
Ethyl,	0·81	10·52	41·7	0·0222
	,,	21·17	64·4	0·0211
	,,	31·38	77·3	0·0205
	,,	41·74	84·0	0·0191
	,,	52·22	88·4	0·0179
Ethyl,	4·81	9·71	32·1	0·0173
	,,	19·51	55·1	0·0178
	,,	29·65	72·2	0·0187
	,,	39·39	82·2	0·0190
	,,	49·31	88·1	0·0187
	,,	59·29	91·0	0·0176

TABLE II.

Alcohol. Initial volume = 200 c.c.	Concentration in grams of Solute per 100 c.c.	$\frac{1}{v}\log\frac{a}{a-x}$
Propyl,	2·5	0·026
n-Butyl,	4·9	0·030
iso-Butyl,	3·4	0·047
sec-Butyl,	2·8	0·050
iso-Amyl,	2·1	0·053

The distillation coefficient $\frac{1}{v}\log\frac{a}{a-x}$, as previously shown, is equal to $\frac{k\sigma}{\rho V}$, and on the assumption that $\frac{\sigma}{\rho}$ is constant, the coefficient varies as $\frac{k}{V}$,

and is, therefore, inversely proportional to the initial volume of liquid distilled. The distillation coefficient varies (but to a less extent) with the concentration, and, therefore, in indicating the values of $\frac{1}{v} \log \frac{a}{a-x}$, it is necessary to specify the initial volume distilled and also the concentration. The values recorded in Tables I and II have initial volumes of 200 c.c., this volume having been found to be a convenient one to work with in the case of the alcohols. As, however, in the work on the fatty acids volumes of 150 c.c., were employed, the values of the coefficient obtained on the distillation of 150 c.c. of the alcohols are given in Table III for comparison.

TABLE III.

Alcohol (V = 150 c.c.)	$\frac{1}{v} \log \frac{a}{a-x}$	Concentration.
Methyl, .	0·015	4·3
Ethyl, .	0·024	4·7
n-Butyl, .	0·040	4·8
iso-Butyl, .	0·066	2·9
sec-Butyl, .	0·066	2·8
iso-Amyl, .	0·072	2·1

Even with the variation in concentration, it will be seen that the results of the two series of experiments at 150 c.c. and 200 c.c. initial volume, confirm that the distillation figures are inversely proportional to the volume.

Comparison of Values for the Coefficient of Enrichment K (Sorel).

The values of $\frac{1}{v} \log \frac{a}{a-x}$ for ethyl alcohol have already (see above) been calculated for two concentrations of the alcohol (0·81 per cent. and 4·81 per cent.), and an appreciable difference is observed between the two sets of results. In order to correlate the variation in the distillation coefficient with the coefficient of enrichment, a more extended series of solutions was studied. The concentrations of ethyl alcohol varied between 4·38 per cent. (by weight) and 0·24 per cent. The values of $\frac{1}{v} \log \frac{a}{a-x}$ for each concentration were determined, and the corresponding values of K calculated. The values obtained are recorded in Table IV.

The values of K obtained experimentally by Sorel varied from 7·15 at 5 per cent. concentration to 9·9 at 1 per cent. concentration. The calculated values of K agree approximately with those obtained directly (see Table IV), thus confirming the relation between the two coefficients we have already deduced.

It will be seen that the distillation coefficients of the alcohols increase in an approximately regular manner with an increase in the molecular weight. The occurrence of a branched chain in the compound results in an increased volatility, as shown by a comparison of the distillation results of the butyl alcohols. The trend of the figures is analogous to that in the case of the lower fatty acids. The fact that the solute of lower boiling point exerts a greater pressure in solution than one of higher boiling point is probably dependent on the fact that the attraction of the unlike molecules (alcohol for water) diminishes with rise of molecular weight of the alcohols, as shown by decreasing miscibility.

In connection with these comparisons it should be mentioned that there is often an evolution of heat and a volume change on mixing the alcohols with water. This is true, however, with large excess of water; but, for example, with sixty molecules of alcohol to forty of water there is an absorption of heat in the case of tertiary butyl, *n*-propyl, and *iso*-butyl alcohols. The contractions on mixing also diminish with rise of weight of alcohol (Young and Fortey).[1]

Table IV.

Concentration of Ethyl Alcohol.	$\frac{1}{v}\log\frac{a}{a-x}$ observed.	K calculated.
Per cent.		
4·8	·0173	7·96
2·6	·0179	8·23
1·75	·0187	8·60
1·14	·0191	8·79
0·74	·0190	8·74
0·38	·0214	9·27
0·24	·0207	9·50

It is seen from Table IV that the distillation coefficient is influenced by the concentration. This is not due to temperature effects, since it has been found that on calculating the value of K, it only diminishes slightly with increasing temperature, as in Table V.

[1] Trans. Chem. Soc., 1902, 81, 717.

TABLE V.

Concentration of Solution.	Temperature.	*K*
Per cent. 80·0	39·76	1·084
80·0	54·8	1·078
80·0	74·79	1·076
60·0	39·76	1·36
60·0	54·8	1·34
60·0	74·79	1·33

The values of *K* in Table V have been calculated from figures given by Vrevsky.[1] The value of *K*, and consequently all the distillation values, are dependent on the concentration of the solution—the greater the concentration the smaller the distillation coefficient. For aqueous solutions of the soluble alcohols of concentration less than that of the constant boiling point mixture the distillation figure increases with increasing dilution, reaching a maximum value in dilute solution. In very dilute solutions the values for the different alcohols are of the order of those recorded in Tables I and II. A comparison of these coefficients with vapour pressures at 100° of the various alcohols shows that there is no apparent agreement between the rates of distillation in aqueous solutions and the volatility of the alcohols.

Although these substances are associated in the pure state, it is improbable that the molecular complexes are not dissociated in very dilute solution. The lack of agreement between the rates of distillation of the aqueous alcohol mixtures and the volatility of the pure alcohol cannot be due to the presence of molecular association of the solute.

There is the possibility that soluble alcohols become associated in part with some of the solvent when dissolved in water; and, if it be assumed that only the free solute is volatile, the concentration in the vapour phase will be proportional to the concentration of the free substance in the liquid phase.

The evidence in favour of the associating theory is, however, very small in these cases. Against the hypothesis that there is any kind of combination or association of the alcohol with water, we have the fact that when the azeotropic mixture of *n*-propyl alcohol and water is cooled in a freezing mixture pure ice separates out. In addition, the formation of an azeotropic

[1] Zeit. phys. chem., 1912, 81, 1.

mixture of minimum boiling point indicates feeble attraction between the unlike molecules.

A more likely explanation may, however, be suggested to explain the results obtained. It is probable that the changes of concentration may bring variations in the "mass of water per c.c. in distillate." Any variation in this quantity will cause A to vary, even though Nernst's law still holds; we have

$$A = \frac{1}{\text{volume distilled}} \log_{10} \frac{\text{initial amount of alcohol}}{\text{amount of alcohol distilled}},$$

and k is related to A thus—

$$A \times \text{constant volume in flask} \times 2{\cdot}3026 = k \frac{\text{mass of water per c.c. in distillate}}{\text{density of water vapour in flask}}.$$

If variations exist, it might be more satisfactory—instead of calculating from the total distillate as shown in Table I—to calculate A from each separate fraction. This we can do, since the volume is kept constant. Table VI gives the new results then obtained.

TABLE VI.

Initial mass of Ethyl Alcohol in grams per 100 c.c.	Weight of distillate.	Mass of water per c.c.	$\frac{A}{\sigma}$
4·81	8·71	0·795	0·0218
	10·81	0·860	0·0208
	10·12	0·905	0·0206
	9·74	0·945	0·0202
	9·92	0·945	0·0202
	9·98	0·945	0·0184

The mass of water per c.c. has been calculated from our analyses and from density tables. The value of $\frac{A}{\sigma}$, it will be observed after correction, gives practically the same value as for dilute solutions, for which σ = 1 approx.

If we take the mean values, omitting first and last determinations, we get $\frac{A}{\sigma}$,

first set (0·81) . . . . 0·0208,
second set (4·81) . . . . 0·0204.

The variation in the weight of water per c.c. of distillate appears to account for the variation in A with concentration within the limits of experimental error; $\frac{A}{\sigma}$ gives the limiting value of A for very dilute solutions.

Preparation of n-Butyl-n-butyrate.

In the estimation of *n*-butyl alcohol with potassium dichromate and sulphuric acid, it was previously noted that some ester was formed when the oxidation was carried out under certain conditions. The conditions which facilitate the formation of this ester were studied as a separate problem.

n-Butyl alcohol (148 grams) was added to a solution of potassium dichromate (200 grams) in sulphuric acid (276 grams in 750 grams of water). Precautions were taken during the addition of the alcohol to prevent the temperature from rising above 30–40°, otherwise the reaction would tend to become vigorous, especially when the alcohol is added too quickly. When the addition was complete, the reaction-mixture was heated for 2–6 hours on a water bath. If the oxidation proceeded satisfactorily, a light-coloured oil was obtained, with a quantity of dark-green lower aqueous layer.

The oil consisted chiefly of *n*-butyl-*n*-butyrate, together with a small amount of unchanged alcohol and products boiling above 190°, and usually a trace of butylaldehyde. In the best experiments a 75 per cent. yield of *n*-butyl-*n*-butyrate was obtained on the original weight of alcohol taken. During some of the experiments, notably those in which the duration of heating was short, the ester layer was contaminated with a green substance. This substance contained chromium, and appeared to be slightly volatile in the vapour of *n*-butyl-*n*-butyrate, for, on distillation of the crude ester, the main fraction was coloured green. It was obtained in a crude state by removing the volatile portions from the residue remaining in the flask after distillation. It was a dark-green amorphous mass, readily soluble in acetone.

As a product of the reaction there was always present a small amount of a substance of higher boiling point than *n*-butyl-*n*-butyrate. The yield of the substance was found to be increased by allowing the oxidation to be completed at laboratory temperature.

The formation of the *n*-butyl-*n*-butyrate may be explained by assuming the intermediate formation of butyric acid which is esterfied by the alcohol. Butyric acid, in fact, has been detected from the aqueous layer, and is usually a product of the reaction.

The following gives a typical example, showing the yields obtained :—

From 150 grams of *n*-butyl alcohol by oxidation the product had the following composition :—

n-Butyl alcohol . .	17·5 grams.
n-Butyl-*n*-butyrate . .	109·4 „
High boiling-point fraction .	3·0 „
n-Butyric acid . . .	8·0 „

The amount of ester obtained is approximately 75 per cent. of the theoretical yield calculated on the alcohol taken.

The ester was identified by hydrolysis yielding *n*-butyric acid and *n*-butyl alcohol. The boiling point of the ester is 166·7°/761 mm.

The authors wish to state that they are much indebted to both Professor Sydney Young, F.R.S., and Dr. F. E. Hackett for many suggestions and useful criticism. They also take this opportunity of expressing their thanks to Captain Desborough for his interest in this work, and for securing permission for publication.

XXII.

THE "BROWNING" AND "STEM-BREAK" DISEASE OF CULTIVATED FLAX (*LINUM USITATISSIMUM*), CAUSED BY *POLYSPORA LINI* n. gen. et sp.

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[PLATES VIII–X.]

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I.—INTRODUCTION.

DURING the recent European war a heavy demand for flax fibre from home-grown sources arose, consequent on the cutting off of foreign supplies. Since, owing to the high prices for fibre then prevailing, successful cultivation of flax was highly remunerative to the farmer, it is not surprising that a boom in flax cultivation set in, so that an immediate and large increase in the area under this crop in Ireland resulted.

This was accompanied in not a few instances by partial or total failure of the crop, which on inquiry and investigation often proved to be due to such causes as unsuitability of soil and unsatisfactory methods of cultivation and manuring, as well as to the use of inferior seed.

In a considerable number of cases, however, it appeared evident that specific diseases caused by parasitic organisms were responsible for serious trouble, and the insufficiency of our knowledge of flax diseases became acutely felt.

Stimulated by this lack of information, a programme for the scientific investigation of flax diseases was forthwith embarked upon by the Seeds and Plant Disease Division of the Irish Department of Agriculture.

A general account of the work undertaken and of the results so far obtained has recently appeared in the Department's Journal,[1] and a scientific

[1] Pethybridge, G. H., and H. A. Lafferty, "Investigations on Flax Diseases," Jour. Dept. Agric. and Tech. Inst. Ireland, xx, 3, 1920, p. 325. See also xxi, 2, 1921, p. 167.

paper dealing with one new specific disease, "Seedling Blight," has also been published.[1]

The present paper deals with another previously unrecognized disease of flax which has been proved to be caused by a fungus hitherto undescribed, and in which a general "browning" of the crop, accompanied by the fracture of many of the stems of the plants, are characteristic features. Up to the present this disease has not been described as occurring in any other country. Nevertheless, there is reason for believing that it must occur elsewhere; and it seems not improbable, from a preliminary note on a disease of flax in Holland, provisionally ascribed to a species of Gloeosporium, published in 1914, that the case in question was one of "browning."[2] The disease occurs in Ireland on flax grown from Dutch seed rather more frequently than on crops grown from seed from other countries. The fungus causing it has been found on seed derived from crops grown in England and Scotland and on seed believed to have been raised in Belgium. The disease has appeared in Ireland on crops from seed purporting to have been raised in England, Scotland, Holland, Russia, Canada, and Japan, and it is believed to be present in British East Africa.

II.—Symptoms of the Disease.

As stated above, the disease manifests itself in two principal forms, and for these the popular terms "browning" and "stem-break" are suggested as suitable.

1. *General features*—(*a*) "*Browning.*"[3]—To the casual observer "browning" first becomes evident about pulling time. The actual time depends to a large extent upon the earliness or lateness of the particular season, but in normal years it is usually towards the end of July or the beginning of August. At this time isolated areas in the crop, of various shapes and sizes, take on a distinct brown colour, so that they contrast somewhat strongly with the still green surrounding plants. Gradually these patches increase in extent until they coalesce, and the entire field becomes more or less uniformly brown. Occasionally, when primary infection is more general from the start, "browning" develops in a more or less uniform fashion over the whole crop, instead of appearing first in isolated areas.

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[1] Pethybridge, G. H., and H. A. Lafferty, "A Disease of Flax Seedlings caused by a Species of Colletotrichum and transmitted by infected seed," Sci. Proc. Roy. Dub. Soc., xv. (N. S.), 30, 1918, p. 359.

[2] Meded. v. d. Rijks Hoogere Land-, Tuin- en Boschbouwschool, Wageningen, vii, 1914, p. 53.

[3] "Browning" is sometimes, but erroneously, referred to as "firing." True "firing" is a very different disease, and is caused by *Melampsora lini*, one of the rust fungi.

The rate of development of "browning" is intimately connected with the weather conditions prevailing at the time, being more rapid in damp warm weather than under relatively dry atmospheric conditions. One diseased centre, approximately one square foot in area, kept under observation, was found to have increased in three days to an irregular area of about seventy-two square feet. Some farmers have long recognized the epidemic nature of the trouble, and they endeavour to pull the crop as soon as "browning" makes its appearance

When individual plants collected from a "browned" patch are carefully examined, it is seen that the fruits, sepals, leaves, and stems show brown diseased areas on their surfaces; and it is to the effect of these when seen in mass that the general browning of the crop is attributable.

On the still partially green leaves the diseased spots, which are generally rounded in outline, may occur either singly or in considerable numbers, as many as eight having been counted on a single leaf. They are dark brown in colour, and, as a rule, sharply marked off from the surrounding tissues, while they are slightly depressed below the level of the normal leaf-surface. As the spots do not increase in size very rapidly, the affected leaves remain only partially attacked, and are able to continue their functions to a considerable extent for some time after infection. In those cases, however, where a leaf becomes attacked in the region of its petiole, death of the whole leaf quickly ensues. In all cases, irrespective of the region of attack, an affected leaf dies prematurely, turns brown, and either falls off the plant or remains with its surface adhering to the stem.

On the main stem the diseased areas are often closely associated with the presence of a diseased leaf situated at a slightly higher level. They are first apparent as minute elongated spots, which are light fawn to brown in colour. Gradually the spots increase in size, especially in a longitudinal direction, and they may even coalesce, laterally as well as longitudinally, until practically the entire surface of the stem is involved (Plate VIII, fig. 2).

In dry weather the cortex often ruptures lengthways along a diseased area, and the stem, when bent at right angles at such a place, has a strong tendency to break; such a brittle condition is, of course, quite foreign to healthy flax.

The attack on the branches is similar to that on the main stem; but, owing to the smaller diameters of the former, the diseased areas more frequently encircle them completely.

Diseased sepals turn brown, and generally adhere firmly to the wall of the fruit.

Affected fruits are easily identified in the earlier stages of attack while

they are still green, but as they ripen they lose their green colour naturally, and become golden or light brown in colour. At this stage the recognition of the disease becomes almost impossible without the aid of the microscope. On the green fruits the diseased areas may arise at any point, but they are generally located at the apex or along one of the five raised ribs. They are, at first, light fawn in colour, and often surrounded by a darker zone due to the development of anthocyan. Gradually, as the disease advances, the darker zone becomes involved, and the colour of each area may then vary from fawn to brown. Fruits showing various stages of attack are illustrated in the two upper rows of fig. 3, Plate VIII. Whether infection originates at the apex or along a ridge, the diseased tissues lose moisture and contract, with the result that affected fruits show a strong tendency to gape open, especially in very dry weather.

As the fruits become ripe and dry, the progress of the disease is retarded; hence many of them show infection on one side only, or on a restricted area about the apex. But in cases of early infection the entire fruit may become diseased, and, if opened, the young seeds within are found to be shrivelled and dead. In cases of slight infection the seeds may appear normal to the naked eye; but, as will be shown later, the majority of them, though still retaining their vitality, are, nevertheless, heavily infected with the disease.

(*b*) *Stem-break.*—In a crop suffering from "browning" there are to be found individuals here and there which, instead of standing upright like the majority of their neighbours, have their stems bent over and lying on the ground. The recumbent position of these stems is due to the fact that at or in the region of the first node they are diseased, and have, therefore, become more or less completely broken across (Plate VIII, fig. 5).

When the fracture is practically a complete one, the plants turn yellow, die, and finally become brown. If, however, the stems are already fairly well developed before the attack becomes severe, and if, therefore, a considerable development of woody tissue has already occurred, only partial fracture occurs. The stems bend over, but since the conducting tissues are not entirely severed, the portions above the breaking-point continue to live and grow for a time. They develop negatively geotropic curvatures, as is shown in Plate IX, fig. 1, but ultimately they succumb prematurely. The connexion between "stem-break" and "browning" was not at first recognized; but, as will be explained later, it has now been proved that these two dissimilar phenomena are due to one and the same cause.

2. *Histological features.*—Microscopical preparations of the tissues from any of the affected parts of flax plants show the presence of hyaline, septate, and branched fungus hyphae in the diseased tissues. The cell contents are

brown and disorganized, while the chlorophyll granules are destroyed. Cross-sections of flax stems in the early stages of the disease show fungus hyphae permeating the parenchymatous tissues of the cortex and travelling in the cells between the fibre bundles. Fungus hyphae have never been observed penetrating the fibre bundles themselves or extending into the wood. A cross-section through a partially diseased stem is shown in Plate VIII, fig. 4.

In such cross-sections the individual fibres of the bundles running through a diseased portion of the stem usually have somewhat thinner walls and larger lumina than those of normal fibres in a healthy region of the stem. Owing to the shrinkage consequent on the death of the epidermal and cortical cells, the fibres are somewhat flattened tangentially.

Micro-chemical tests show that the fibre bundles in a diseased region undergo certain changes in composition. Thus, with Schulze's solution, the thickened parts of the fibre walls slowly become violet in colour, while the middle lamellae are stained yellow. The walls of normal fibres when treated with this reagent quickly become blue, while the middle lamellae remain undifferentiated. When cross-sections are treated with concentrated sulphuric acid, the thickened walls of the fibres in a diseased region swell up and become dissolved; but the middle lamellae remain insoluble, and present the appearance of a brown cellular skeleton. In a healthy region the middle lamellae also disappear. With a solution of iodine in potassium iodide the middle lamellae of the fibre bundles in a diseased region are coloured yellow, while the thicker portions of the fibre walls remain unstained. Phloroglucin stains the wood a bright red in all regions; but in a diseased region it also stains the middle lamellae of the fibre bundles red, whilst the walls of the individual fibres themselves are also to some extent reddened. This does not occur with normal fibre bundles; and the reactions suggest that lignification of the walls of the fibres has to some extent occurred. Correlated with this change is, doubtless, the fact already alluded to, that at diseased regions the stems become somewhat brittle.

Since, as will be seen later, the disease is one which is transmitted by means of the seed, special attention was paid to the manner in which the seeds become infected with the fungus. For this purpose, fruits at all stages of development were collected and studied microscopically by means of cross-sections. The progress of the mycelium was traced from the outside of the fruit through the tissues of its walls to the placenta, until it reached the seed itself by traversing the extremely short funicle. If the seeds are young, and the several layers of the seed-coat consequently not fully developed, the fungus penetrates these tissues, and reaches the young embryo, which becomes

permeated with mycelium, and is killed. Such dead seeds eventually become covered externally with conidia, and, as a rule, remain adherent to the inner walls of the fruit.

If the seeds are more nearly ripe before infection of the fruit occurs, the fungus, as before, reaches the two outer layers of the seed-coat, but does not penetrate the third or fibrous layer. Hence the embryo remains secure from attack, and its subsequent development proceeds normally. Although unable to gain entrance to the embryo, the fungus often continues to spread in the outer layers of the seed-coat, and produces conidiophores and conidia on the surface of the seed.

As the infected seeds become ripe they break away from the placenta, and the fungus produces conidiophores and conidia in great abundance on the resulting scar. Slightly affected seeds show the presence of conidia only at or in the region of the hilum.

3. *Details of the Fungus.*—When portions of affected plants, whether stems, leaves, sepals, or fruits, are kept for forty-eight hours or so in a moist atmosphere, minute beehive-shaped pustules (*acervuli*), scarcely visible to the naked eye, are produced on and around the margins of the discoloured diseased areas.

These acervuli, which occur, as a rule, directly over the stomata, are gelatinous in consistency and hyaline to milky in appearance. They are made up of large numbers of conidia, produced by groups of conidiophores, the swollen tips of which emerge only slightly through the stomata. One such conidiophore emerging on the surface of a diseased leaf and producing conidia is shown in Plate X, fig. 5.

Occasionally on diseased stems and branches, especially when the disease has advanced to a very considerable extent, cases are met with where masses of conidia are to be found in minute pockets beneath the epidermis, as well as on its surface. These submerged conidia are produced by conidiophores which at first produced conidia in the sub-stomatal respiratory cavity. Eventually, however, owing to the pressure from beneath, the epidermis bursts, and the conidia are forced out in bulk. Conidia have never been observed to be extruded through a stoma in the form of a tendril, as takes place commonly in the case of a pycnidium with an ostiole. This submerged production of conidia has never been observed on leaves, and, though sometimes seen on stems and branches, it is not the usual method. In the majority of cases conidia are produced outside of the host.

The acervuli have been examined carefully in all stages of development, and on all parts of the host plant for the presence of setae, but the latter have never been met with.

The conidia, which have bluntly pointed ends, are hyaline, single-celled, and of varying shape—oval, cylindrical, and straight forms with a peculiar basal twist are those most commonly met with: see fig. 1, Plate X. Stained preparations reveal the fact that the wall of each conidium is slightly thickened at its base, where it was attached to the conidiophore. Their contents are finely granular, and they sometimes possess one or more oil-drops. Though very uniform in breadth, which averages 4μ, they vary greatly in length, the extremes being 9μ and 20μ, with an average of 15μ.

They are produced by abstriction on the swollen ends of the conidiophores, and occasionally for a short distance along the sides of the latter. Each conidiophore may produce from one to seven conidia at a time; but three, four, and five are most often seen. When the conidia are ripe they become detached from the conidiophore, and new ones are then produced. Owing to the adherence of the conidia to one another, a little heap or mound of them is formed where the free ends of the conidiophores emerge.

In the case of the leaf, the conidiophores arise from a loose mass of hyphae situated towards the base of the respiratory cavity beneath a stoma. These hyphae resemble those that permeate the dead tissues of the leaf; but they are not sufficiently numerous or so densely packed as to permit of the aggregation of them formed in the respiratory cavity being regarded as a stroma. The conidiophores continue to grow in the form of an aggregation of rather thick hyaline parallel hyphae towards the stoma, through which their tips eventually emerge. As a rule they are simple, but occasionally they are seen to branch in an irregular manner. On emerging, their free ends, which never protrude very far, immediately become slightly swollen, and they begin to produce conidia. Simple unbranched forms were found to measure on an average 27μ long by $6{\cdot}5\mu$ broad, the latter measurement being made across the broadest part of the swollen apex.

On flax stems the formation of the conidiophores and conidia resembles in general that on the leaf. In some cases, however, especially when the cortex of the stem is in an advanced stage of disorganization, the parenchymatous tissues beneath the epidermis become ruptured, and the cavities thus formed become filled with a more compact mass of hyaline hyphae from which, here and there, conidiophores arise. These, if they have sufficient space, may produce sub-epidermal conidia as previously described, but more frequently they burst out through a stoma and produce conidia on the surface of the stem.

As already pointed out, this somewhat compacted mass of hyphae is by no means constantly associated with attacked stems, and even when present it is not sufficiently well developed to merit the description of a stroma.

The swollen ends of the conidiophores have been very carefully examined for the presence of sterigmata, but these have never been seen. Occasionally very minute local thickenings have been found in the walls at the ends of old empty conidiophores, which may, perhaps, represent points at which conidia were produced, but no general thickening of the wall of the conidiophore has been observed.

III.—Isolation of the Fungus and Behaviour in Pure Cultures.

Owing to the abundance of conidia produced by the fungus, there is no difficulty in obtaining it in pure culture. Such cultures, starting in each case from a single conidium, have been derived from the fungus present on diseased fruits, seeds, leaves, and stems, as well as on the affected tissues of plants exhibiting the "stem-break" phase of the disease. In every case detailed studies proved that the organism obtained was the same. It is easily cultivated and grows reasonably well on a variety of nutrient media.

Diluted conidia-suspensions in wort gelatine, or beef-extract gelatine, were plated out in Petri dishes in the usual way. When the gelatine had solidified, the dishes were examined with the low power of a microscope, and the positions of isolated conidia were marked. When germination had commenced, a single germinating conidium, together with a small piece of the medium surrounding it, was transferred, with aseptic precautions, to each of several tubes of slanted media; hence the growth in each tube was the product of a single conidium.

For purposes of comparison, the fungus isolated from different parts of the host and from flax plants growing in widely different localities in Ireland has been grown on the following media:—beef-extract gelatine, beef-extract agar, extract of Quaker-oat agar, ground Quaker-oat agar, cooked green flax stems and nutrient agar made from the watery extract of cooked green flax stems; but irrespective of its source of origin, the fungus behaved similarly on the various media used, and showed no morphological or cultural differentiation into strains or races.

The fungus develops its mycelium comparatively slowly and sparsely on all the media tried, but conidia are always produced in great abundance. On *beef-extract gelatine* and *agar* growth is poor, the mycelium being, for the most part, submerged. It is hyaline, septate, and branched, the individual cells being thin-walled and irregular in size and shape, and frequently much swollen. The surface of the medium becomes covered with a cream-coloured mass of conidia resembling a bacterial growth. The conidia are somewhat larger on the gelatine medium than they are on the host, their dimensions averaging $15\mu \times 5\mu$. On agar they averaged $14\mu \times 4{\cdot}6\mu$. Many of them

showed the production of new conidia from their ends. The gelatine is slowly liquefied, the process not being complete even after four weeks. Agar is not affected.

Cold water *extract of Quaker-oat agar* is a poor medium for the growth of the fungus. The amount of mycelium produced is relatively small, but dirty white masses of conidia are produced in good amount on the surface of the medium. The conidia averaged $13{\cdot}7\mu \times 4\mu$ in length and breadth respectively; and in shape they closely resembled those produced on the host plant.

Ground Quaker-oat agar was found to be the best medium for long-continued growth. On it the fungus grows slowly, but continues growing until the entire surface of the slant is covered, this being accomplished in about one month. The mycelium is again submerged, and conidia in cream-coloured, slimy masses are produced in abundance on the surface of the medium. After growing for several days, the submerged mycelium gradually becomes dark in colour, and as time goes on this darkening increases in intensity until a dense wrinkled mass of matted black mycelium is produced at or immediately below the surface. When examined microscopically, the dark mycelium is found to consist of much-swollen, more or less spherical thin-walled cells, with finely granular contents. Individually these cells are olive brown in colour, but when seen in mass the effect produced is almost black. The individual swollen cells of this dark mycelium eventually break away from one another, but their subsequent behaviour was not followed. It is believed, however, since they have never been seen in the tissues of the host plant, and are produced only on certain nutrient media, that they have no special significance in the bionomics of the fungus. Occasionally the olive tinge was taken up by the conidia or was diffused into the substance of the medium. As a rule, however, the conidia are hyaline, except where they are in intimate contact with the dark hyphae, and they averaged $14{\cdot}5\mu \times 4{\cdot}1\mu$ in length and breadth respectively.

The fungus grows well on *cooked green flax stems*, and produces acervuli on their surfaces. These acervuli are only just visible to the naked eye as very minute black dots resembling pycnidia. Microscopical examination shows that each conidial mass is in reality hyaline or milky in appearance, but, as it is seated on a dark-coloured base made up of the conidiophores whose swollen apices in this case are olive-coloured, it appears to have a smoky tinge. The conidia in the acervuli are similar to those found on the living host, and average $14{\cdot}3\mu \times 3{\cdot}8\mu$.

The mycelium in the tissues of the cooked flax stems is thick, hyaline, and much septate, the darkening only showing where the conidiophores burst through the epidermis.

Cooked green flax-extract agar is an unsuitable medium for the growth of the fungus; the submerged mycelium is sparse, with a complete absence of discoloration. Slimy, dirty white masses of conidia are produced on the surface of the medium, and, to a lesser degree, conidia are formed in the medium itself. They are very variable in shape and size, but average $15\mu \times 4{\cdot}3\mu$.

On all the media mentioned the conidia are produced in masses from the swollen tips and along the sides of the hyphae, which are little, if at all, differentiated from the ordinary mycelium, and which, therefore, scarcely deserve description as definite conidiophores. On the sides of the hyphae or on surface mycelium they are produced, one or more at a time, on the apices of short, blunt "pegs," which jut out from the cell wall, but which are very distinct from sterigmata; see fig. 6, Plate X. On the ends of the hyphae, which, as a rule, are slightly swollen, the conidia are produced by abstriction, and they may arise at any point; see fig. 3, Plate IX.

It sometimes happens that if, for some reason or other, a cell of the mycelium loses its protoplasm, the neighbouring cell behaves as a conidiophore, and conidia are produced in succession from it, and are pushed into the empty cell. Such a case is shown in fig. 8, Plate X.

The germination of the conidia, taken from the host as well as from pure cultures, has been followed in hanging drops of tap water, of the liquid extract of cooked flax, and also on film cultures of beef-extract gelatine, wort gelatine, and extract of oat agar.

In tap water, before germination, the conidia become much swollen, and in many cases two-celled, by the formation of a transverse septum. This causes a constriction in the middle of each conidium. From either or both ends of the conidia new conidia are produced, but no germ tubes are developed.

Where nutrients are used, however, the conidia swell rapidly, and in almost every case a transverse septum is produced, previous to the emission of a germ tube from one or both ends of the swollen conidium. As a rule these germ tubes soon become branched, and begin to produce new conidia at their ends after they have grown a short distance. Gradually, however, they increase in length by the development of new terminal cells, and these in turn bear conidia at their ends and along their sides until a fungus growth results, with a small amount of mycelium, but an abundance of conidia. The germination of the conidia, their subsequent growth, and the production of new conidia are illustrated in figs. 2, 3, and 4, Plate X. On wort-gelatine film cultures the early form of growth of the fungus is distinctly star-shaped when seen under the microscope, as illustrated in fig. 2, plate IX; but, as growth proceeds, lateral branches are formed on the radiating hyphae, and the star-shaped effect becomes obliterated.

IV.—Description and Position of the Fungus.

During the long period in which the growth and development of the fungus have been studied, both on various media in pure culture and on its natural host, the flax plant, no method of reproduction other than by conidia has been met with. It is clear, therefore, that for the present, at any rate, it must be placed amongst the Fungi Imperfecti.

If emphasis be laid on its mode of development on the host, the formation of conidia in definite acervuli would suggest its inclusion amongst the Melanconiales. On the other hand, judging from its mode of growth as a saprophyte on various media, it might well be placed with the Hyphomycetes.

At the outset the fungus was provisionally ascribed to the genus Gloeosporium, but detailed study shows that it cannot remain there. The figure given by Lindau[1] of the conidiophores of *Microstroma Juglandis* Niessl emerging through a stoma recalls at once the behaviour of the flax fungus. The genus Microstroma has been placed amongst the Exobasidiaceae, but Maire's investigations[2] show that it should rather be classed amongst the Fungi Imperfecti. In any case, however, in this genus the conidiophores arise from a definite submerged stroma, and no such stroma is developed by the flax fungus. As a matter of fact, the characters of the latter do not admit of its being placed in any genus hitherto described; hence it becomes necessary to make it the type of a new one. Since one of its leading features is the production of large quantities of conidia, with relatively little development of mycelium, the name Polyspora is suggested for the genus which will find a place in the Melanconiales. It is characterized as follows:—

Polyspora nov. gen.

Mycelium, hyalinum, septatum, intra hospitem ramosum: basidia ex hyphis laxe in cavo substomatali conglomeratis surgentia ad summum subtumida et per stomata paullum protuberantia simplicia vel subramosa ad summum plura conidia quodque gerentia: acervuli numerosi minuti gelatinosi hyalini vel lactei in hospitis epidermide super stomata raro subter siti: conidia hyalina continua obovata vel cylindracea terminis rotundatis complura simul ad summos vel in lateribus basidiorum gesta.

P. lini. nov. spec. E genere supra descripto; conidiis 9–20μ (medium 15μ) longis 4μ latis nonnumquam cum una vel duabus guttulis.

Hab. in foliis, stirpibus, fructibus seminibusque vivis *Lini usitatissimi* in Hibernia.

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[1] Lindau, G., Rabenh. Krypt. Flora. I, 8, p. 19.

[2] Maire, R., La structure et la position systematique des *Microstroma* et *Helostroma*. Nancy. 1913.

V.—PROOF OF PATHOGENICITY OF THE FUNGUS.

Experiments which proved that Polyspora is pathogenic to flax, and was the cause of "browning" and "stem-break," were carried out under strictly controlled conditions in the greenhouse. Other experiments were carried out in the field, where, of course, less control of the conditions was possible. These infection experiments will now be dealt with.

Two small lots of flax seed, previously proved by microscopical examination to be free from fungus infection, were sown in pots of soil previously heated in an autoclave at 140° C. for two hours on each of three successive days. A perfectly healthy and luxuriant crop of seedlings arose, and the plants were allowed to grow until the petals of the flowers had fallen and the fruits were beginning to form. The plants in one pot, which served as a control, were then sprayed with sterile water and at once covered with a large glass bell-jar. Those in the second pot were sprayed with a suspension in sterile water of conidia from a thirty-day-old pure culture of the fungus, and were likewise covered with a large bell-jar.

The experiment was started in August, and the plants were left in an unheated greenhouse. They were carefully examined daily; but the bell-jars were not removed for this purpose. The first indications that infection had taken place were noticed on the twentieth day after spraying. Circular brown spots appeared on the leaves, and elongated brown areas on the stems and branches, of the plants which were sprayed with the suspension of conidia, thus reproducing the typical symptoms of "browning" as it occurs in the field. After a further lapse of a few days portions of affected leaves, stems, and branches were removed and examined microscopically, when typical acervuli, made up of the conidia and conidiophores of the fungus, were found on the diseased areas in all cases.

The conidia present in these acervuli were plated out, and the fungus obtained was proved by comparative tests in pure cultures to be identical with that from which the conidia used for inoculation purposes were obtained. At no time throughout the experiment did the control plants show any signs of disease whatever.

Two adjacent plots, containing only healthy flax plants, were selected among those on the land attached to the field laboratory. Each plot was four square yards in area, and when pulling time was near at hand the plants in one plot were sprayed with sterile water, while those in the second were sprayed with a suspension in sterile water of conidia from a sixteen-day-old pure culture of the fungus.

After a lapse of sixteen days the plants in the plot sprayed with the conidial suspension had become distinctly brown, and resembled in every detail those naturally affected with the disease. Those in the control plot assumed the normal golden colour characteristic of healthy ripe flax, and showed not the slightest trace of "browning."

The "stem-break" phase of the disease was also artificially reproduced in the following manner:—Two pots of soil were sterilized as before, and flax seeds proved to be fungus-free were sown in each. After sowing, the plots were covered with bell-jars and kept in the greenhouse. When the seedlings were about one and a half inches high all the cotyledons of the plants in one pot were wetted with sterile water, while the cotyledons of those in the second pot were smeared with conidia from a twelve-day-old pure culture of the fungus. The bell-jars were then replaced. During subsequent waterings of the soil care was taken not to wet the cotyledons; consequently the conidia remained undisturbed on them.

The smeared cotyledons showed the first signs of infection seventeen days after the conidia had been applied to them. They gradually turned brown, and, owing to the general infection which took place over their whole surfaces, isolated spots did not develop. The progress of the fungus along the cotyledons was followed until the latter reached the main stems of the plants at the nodes. Here it continued to spread, with the result that the tissues of the stem became diseased and brown; but its further progress was not rapid, and the injury was more or less localized.

Twenty-four days after smearing all the cotyledons were dead, brown, and shrivelled, and one plant had toppled over, the stem being nearly broken through a little below the first node. In this case the disease extended down along the stem for approximately half an inch; but the greatest damage to the tissues occurred where the partial fracture took place.

The experiment was continued for forty-six days, by which time five other plants had fallen over, the partial fracture of the stem being at the first node in each case; see fig. 1, Plate IX. The remaining plants were then carefully examined, and in every case their stems at the point of insertion of the cotyledons were found to be more or less diseased. Owing to the tranquil conditions prevailing underneath the bell-jar, they continued to remain erect, but had they been subjected to the disturbing effects of wind and rain, such as would be the case under open field conditions, many more of them would, in all probability, have fallen over.

The dead cotyledons and the diseased areas on the stems of those plants that had fallen over were examined microscopically, and typical acervuli of the fungus were found in abundance on them. Finally, it was proved by

raising pure cultures from single conidia of the fungus present that the latter was identical with the one used for inoculation purposes.

The plants in the control pot remained healthy throughout the experiment. In many cases their cotyledons lost their green colour and turned yellow, but no fungus appeared on them.

VI.—Disease Transmission.

During the inspection of large-scale field plots of flax derived from seed having different countries of origin, it was early recognized that the plants from certain kinds of seed, notably that imported from Holland, were often heavily attacked by "browning," while crops from seed from other countries, though grown under similar conditions, often actually side by side, were comparatively or quite free from it. Even before it was discovered that the seed itself is liable to attack, this observation strongly suggested that transmission of the disease probably took place in some way by means of the seed.

In several instances samples of the seed from which a diseased crop arose in the field were obtained, and in every case it was definitely proved by microscopical examination and cultural trial that such seed was infected with the fungus.

The method of seed examination for the presence of the fungus was as follows:—A number of drops of water were placed on a microscope slide, and a single seed from the sample under examination was placed in each drop and manipulated until it became thoroughly wetted. The drops were then examined microscopically with a low power. In the first few trials nothing abnormal was noticed. Eventually, however, conidia identical with those of *P. lini* were seen in abundance in the water surrounding one of the seeds; and, as the examination proceeded, numerous examples were found where a similar state of affairs occurred.

In the majority of cases where conidia were seen, the latter were present in masses around the hilum of the seed, and they floated away singly or in groups into the surrounding water. They were, however, by no means confined to this region, often being present over the entire surface of heavily infected seeds.

Several of these seeds on which conidia were present were more closely examined with a view to ascertaining whether the conidia were merely adhering mechanically to their surfaces or were associated with internal mycelium in the seed-coat. Transverse sections of such seeds when examined microscopically showed that the outer layers of cells of the seed-coat were permeated with hyaline, branched and septate hyphae; and the conidia were,

for the most part, formed on conidiophores which sprang from these hyphae and had burst through the outer wall of the epidermal cells. The conidiophores resembled those of *P. lini* as found on browned flax, but they were not grouped together as they occur when protruding through a stoma. They were more or less isolated, and emerged singly and in an irregular manner all over the surface of the seed, so that definite acervuli were not produced. At the hilum the conidiophores were produced in greater quantity than elsewhere. Two simple conidiophores producing conidia on the surface of a seed are shown in fig. 7, Plate X.

As has previously been stated, the fungus hyphae in the seed-coat can clearly be traced in the epidermal and underlying parenchymatous tissues as far as the outer edge of the fibrous cells, but they have never been seen to penetrate the latter.

Occasionally masses of conidia have been found completely filling individual cells of the dead parenchymatous tissue beneath the epidermis, and in some cases they are formed in the epidermal cells themselves. Generally speaking, however, they are produced on the exterior of the seed-coat.

Owing to the rupture of the cuticle and walls of the epidermal cells by the conidiophores of the fungus, affected seeds absorb water more rapidly than healthy ones do; consequently such seeds become mucilaginous more quickly, and this fact is an aid to their detection.

Trials were made to ascertain whether the conidia present on the seed-coat were alive, and, if so, whether they had any connexion with the outbreak of "browning" on the plants in the field. They were found to be alive, and by the ordinary poured plate method pure cultures (starting in every case from a single conidium from the surface of an affected seed) were raised. Further, by comparative cultural tests the fungus so obtained was proved to be identical with that isolated by means of the conidia taken from browned stems, leaves, and fruits.

As a result of microscopical examination, fifty-two seeds bearing conidia were picked out from a sample which had produced a browned crop the previous year. These seeds were sown approximately one inch deep in a pot of sterilized soil, and after sowing the pot was covered with a bell-jar and kept in an unheated greenhouse. Since the seeds were sown in October, germination was slow, and the plants, which were examined daily, made poor growth.

The first sign of disease was noticed on the thirtieth day after sowing. The cotyledons of several of the seedlings showed little brown rounded areas, which gradually increased in size and became dark in colour. The experiment

was concluded on the thirty-third day after planting, and out of a total of forty-six plants fourteen showed diseased cotyledons. As the seedlings had not been watered, and the bell-jar was not removed before the appearance of the disease on the cotyledons, their infection must have resulted from the fungus present on the seeds sown.

Diseased cotyledons from the infected seedlings were examined microscopically, and it was found that the brown tissues were filled with hyaline, septate, and branched hyphae, while groups of conidiophores were bursting out through the stomata and producing conidia in typical acervuli of *P. lini* on the surface. Isolations of the fungus, starting in each case from a single conidium from one of these acervuli, were made, and in every case it proved to be identical with the species found on the leaves, stems, fruits, and seeds of browned plants. This experiment shows that the disease is capable of transmission by means of infected seed.

As is well known, flax seeds when sown and lightly covered with soil germinate, and in a great many cases their seed-coats are carried above soil-level, attached more or less loosely to the cotyledons. Where infected seeds are sown it is obvious that the infection of the cotyledons may take place from the fungus present on the seed-coats borne aloft in the manner described. In the experiment described above a record was not kept of the number of seedlings which carried up their seed-coats, but, as the seeds were planted approximately one inch deep, the number must have been small.

In the spring of 1919 flax seeds from a sample which had produced a browned crop in 1918, and some of which were proved by microscopical examination to be infected, were sown out of doors in a plot attached to the field laboratory.

When the plants had brairded, many of the cotyledons of the seedlings showed the presence of one or more brown circular spots, which on incubation in a moist atmosphere produced acervuli typical of the "browning" fungus. From conidia present in the acervuli, *P. lini* was isolated and grown in pure cultures. Such diseased areas on the cotyledons, some of which are illustrated in fig. 1, Plate VIII, were thus found to be the incipient stage of "browning" as it occurs in the field.

Cotyledons attacked by *P. lini* resemble very closely those attacked with "seedling blight," and in some cases the two diseases at this stage can only be differentiated with the aid of a microscope. To the naked eye the injury due to the "seedling blight" fungus (*Colletotrichum linicolum*) is lighter in colour than that due to *P. lini*, and its progress is also more rapid. In the former case an infected leaf soon becomes entirely destroyed; and this is often followed by death of the seedling as a whole, owing to the formation of

stem lesions in the region of the soil-level which cause the plant to "damp off." In cases of attack by *P. lini* the affected spots on the cotyledons are darker in colour, and do not increase in area rapidly, so that the cotyledons remain alive for a considerable time after infection has taken place. True, as mentioned in an earlier part of this paper, lesions due to *P. lini* have been found on the stems of affected plants in the region of the first node, as a result of which the latter fall over; but this condition of things generally occurs at a later stage, when the flax is at least six inches high, and it in no way resembles "damping off," as found in "seedling blight."

When there is any doubt as to which organism is at work the matter can easily be settled by microscopical examination of the acervuli present on the diseased areas. If *P. lini* is responsible, these acervuli will agree with those herein described; but if *C. linicolum* is the active parasite, the acervuli will be found to be made up of conidia borne singly in succession on conidiophores, while from the bases of the acervuli long, black, tapering setae arise.

As the seedlings affected with *P. lini* grew in size, some of the new leaves produced became infected through various agencies, from conidia present on the diseased cotyledons. From these leaves, in turn, adjoining plants became infected, with the result that when the crop was half grown the disease was general, though not very obvious. Some plants exhibited the "stem-break" form of injury, and were either dead or dying, while the leaves of others were spotted, and in a few cases minute elongated brown marks were making their appearance on the stems.

This state of affairs persisted until after the plants had flowered and the fruits were beginning to be formed. Then the fungus became more virulent, possibly owing to the decreased vitality of the plants or to some change in the environmental conditions, and "browning" of the leaves, stems, branches, and fruits became most pronounced. Thus it is seen that the fungus which causes "browning" is present, though not to a large extent, on some of the flax plants, at least from a very early stage in their development.

VII.—Control of the Disease.

The losses due to "browning" and "stem-break" are, in many cases, very serious, although naturally it is a difficult matter to estimate them quantitatively with accuracy.

"Browned" stems tend to break in the regions of the diseased areas during scutching. Thus the fibre is shortened, and much of it escapes with the tow. Further, plants affected with "stem-break" may be dead and useless at pulling time, and, in any case, are most likely to be left behind when the crop is pulled.

The yield of seed from a "browned" crop is a reduced one, and, of course, its quality is inferior. In Ireland, generally speaking, no attempt is made to save flax seed for sowing, and such loss may, perhaps, be regarded as of no serious consequence. Nevertheless, the work of the Irish Department's Plant Breeding Division in recent years has clearly shown that high-class fibre-flax seed can be produced at home; and the experience of the late war shows the great risks involved in depending on foreign sources alone for the supply of flax seed. Hence the desirability of endeavouring to find some means of controlling the disease.

Persistence of the Fungus.—Polyspora lini, as already stated, has not been found to produce any reproductive bodies other than the thin-walled conidia already described. It has been found, nevertheless, that this fungus can remain alive in a dormant condition in the host tissues for considerable periods, and under seemingly adverse conditions. Thus, flax stems affected with "browning" and containing the fungus were pulled in July, 1920, and allowed to become air-dry in the laboratory. After being subject to these conditions for six months they were exposed to a moist, warm atmosphere in a covered glass dish. In the course of a day or so the fungus recovered its activities. Large numbers of new acervuli were produced on the surfaces of the stems; and the newly formed conidia were found to germinate normally when transferred to films of beef-extract gelatine.

Affected stems pulled a year earlier than those just mentioned, and kept under similar conditions for eighteen months, showed a very feeble development of acervuli when placed in a moist, warm atmosphere; and only a scanty development of conidia took place. Nevertheless, a few of the latter were found to be viable.

In the ordinary course of events, affected stems after pulling go into the retting pond; but what happens to the fungus under these circumstances has not yet been fully ascertained. Stems affected with "browning," pulled and retted in 1919, were air-dried immediately afterwards, and were kept in the laboratory in this condition for eighteen months. When placed in a moist, warm atmosphere at the end of this period, further growth of the fungus and development of new conidia could not be detected with certainty.

No experiments have as yet been carried out with affected stems, either retted or non-retted, exposed to ordinary outdoor weather conditions over the winter. From what has been said above, however, it would appear possible for the fungus to remain alive in affected flax stems over the winter, and for such stems to form starting-points for the infection of the crop in the following season. That the latter is actually the case, however, seems highly improbable.

In Ireland flax is always grown in rotation with other crops, so that on a

given field a second crop of flax normally follows a previous one only after an interval of six or seven years. It seems scarcely likely that affected stalks, left lying in the field after the first crop, could possibly be a source of infection in such circumstances. It is more probable that they would have disappeared entirely and the fungus with them. There is no evidence at present, at any rate, that the fungus can live as a saprophyte in the soil; nor is there any evidence that infection of the crop comes from the soil. Again, after retting, flax is spread on grass fields to dry, but such grass land is not in practice laid down to a crop of flax in the following season, and often not for some years. Even if a few affected stray stalks were left on such land, and assuming that the fungus was not materially injured during the retting process, it seems improbable that infection would occur from this source.

The "shives" (débris consisting of woody cylinder and cortex) which accumulate at scutching mills, where the fibre from the retted flax is mechanically separated from the rest of the stem, are sometimes used for bedding farm stock. It is conceivable that should the fungus survive the retting process it might ultimately reach the land again in manure derived from such material. It is believed, however, that even if it exists at all, this possible source of infection is of no practical significance; and, as has been already stated, there is no evidence at present that the disease is contracted from the soil.

Ensuring Healthy Seed.—On the other hand, it has conclusively been proved that "browning" and "stem-break" can be transmitted by sowing infected seed;[1] and it is believed that this is the most important, if not the only, means by which outbreaks of the disease arise. Control measures, therefore, centre around the question of the seed; and the most obvious precaution would be to avoid saving seed for sowing from a crop suffering from "browning."

It might be considered possible to save healthy seed from a diseased crop by pulling the latter before the fruits and seeds had become infected. An attempt was, in fact, made to do this, but, owing to the early date at which pulling was necessary, the quantity of seed saved was extremely small; and it was found to be altogether too immature to be of any use for sowing purposes.

Again, it might be thought possible to check "browning" and prevent infection of the fruits by spraying the affected crop with an appropriate fungicide. It might, indeed, be feasible to spray the marginal portions of a field of flax at the time when "browning" was beginning to become evident,

[1] Viability tests of the conidia present on infected seeds showed that they retained their vitality on the dry seeds for two years and six months after harvesting. In three years, however, the conidia were all dead. The germination of the seed in this period had, of course, greatly diminished.

in so far as they could be reached from the headlands; and such portions might possibly be sufficiently large to provide the requisite quantity of seed in a given case. But until it can be done from the air by a hovering aeroplane, or by a dirigible airship, or other such means, spraying a flax crop at such a time will not be a practicable proposition, since with present-day machinery the injury which would follow the trampling down of the crop would be irreparable. In addition to this, there is to be considered the effect which the adhering fungicide might have on the subsequent process of retting. Whether spraying at a much earlier stage would be feasible without injury, and, if so, whether it would be effective in preventing "browning," is a problem which has yet to be explored.

Mechanical Avoidance of Infection from Seed-coats.—Since primary infection occurs in the seedling stage, and is due to the transfer of the parasite from the infected seed-coats to the cotyledons, on which the former are borne above the ground, it might be thought possible to intercept such primary infection by preventing the carrying aloft of the seed-coats; that is, by deep sowing of the seed.

Healthy seed was sown in plots at depths varying from half an inch to three inches; but it was found that sowing at a greater depth than one inch resulted in the production of such a thin braird of weak seedlings that a reasonable crop was quite out of the question. Moreover, it was also found that sowing infected seed at a depth of one inch did not suffice in many cases to prevent the seed-coats being carried up on the cotyledons, and infection of the seedlings was found to occur under such conditions. Hence, control of the disease by deep sowing of the seed is out of the question from a practical point of view.

Seed Disinfection.—Considerable attention has been devoted to the question of disinfecting affected seed both by means of fungicides and by heat; and a summary of what has been done along these lines will now be given. At the outset it may be remarked that flax presents special difficulties in regard to treatment with most fungicides in solution or suspension in water, owing to the mucilaginous nature of the outermost layer of the seed-coat, and the tendency of the treated seeds to adhere together in a sticky mass when wetted, thus rendering sowing extremely difficult or quite impossible. Experiments are in progress with a view to finding a way out of this difficulty, but they have not yet been carried sufficiently far to warrant discussion here.

Preliminary trials were made with aqueous solutions of copper sulphate and mercuric chloride, as well as with Burgundy mixtures (copper sulphate and sodium carbonate solutions) of varying strengths. Formaldehyde both in aqueous solution and in gaseous form was also tried. Sufficient seed to sow

an area of about twenty-five square yards was treated in each case, and the plots were situated close to the field laboratory at Boghill.

Copper Sulphate.—Affected seeds were lightly sprayed with an atomiser with 5 per cent. and 10 per cent. solutions of copper sulphate respectively. After spraying the seeds were spread out and dried for twenty-four hours at 40°C. The treatment had no adverse effect on the germination of the seed when tested in the usual way. The plants arising from the treated seeds when sown in the field, however, were not free from the disease.

Mercuric Chloride.—Similar trials were made with 0·1 per cent., 0·5 per cent., and 1 per cent. aqueous solutions of mercuric chloride, and the percentage of germination of the treated seed was found not to be appreciably lowered. Seedlings arising from seed treated with the 0·1 per cent. solution were, however, seriously diseased. The stronger solutions were more efficacious in checking the disease, but neither of them suppressed it entirely.

Burgundy Mixtures.—Affected seeds were sprayed with Burgundy mixtures of different strengths made by neutralizing 2 per cent., 10 per cent., and 15 per cent. aqueous solutions of copper sulphate with the requisite quantities of sodium carbonate solutions. The seeds were then dried, and the adhering blue precipitate remained as a coating on their surfaces. Germination tests showed that the vitality of the seed was in no way injured by the treatment. It was found, however, that the plants derived from such treated seed were not less affected with "browning" and "stem-break" than were those derived from untreated seed sown in an adjoining control plot.

Formaldehyde Solution: Steeping.—Affected seeds were steeped (with occasional shakings to ensure thorough wetting) for periods of five and ten minutes in weak aqueous solutions of formaldehyde having strengths of 0·09 per cent. and 0·18 per cent. respectively. After steeping, the seeds were separated from one another, placed on absorbent paper, and then dried for forty-eight hours at 35°C. Tests were made before and after treatment both of the germination of the seeds and of the viability of the conidia of the fungus. The results are summarized in the following table :—

Strength of Solution.	Time of Steeping.	Germination of	
		Seed.	Conidia.
Untreated.	—	94 per cent.	Abundant.
0·09 per cent.	5 minutes.	94 ,,	,,
0·09 ,,	10 ,,	96 ,,	Nil.
0·18 ,,	5 ,,	80 ,,	,,
0·18 ,,	10 ,,	46 ,,	,,

Steeping in the stronger solution satisfactorily disinfected the seed, but retarded and seriously reduced its percentage of germination. Steeping for ten minutes in the weaker solution killed the fungus, and did not reduce the germination of the seed, while steeping for five minutes only in this solution did not kill the fungus.

A repetition of the experiments gave similar results, with the exception that steeping for ten minutes in the 0·09 per cent. solution did not kill all the conidia; a few were found still viable after treatment.

Owing to the sticking together of the steeped seeds, however, it was found difficult to treat a quantity sufficient for sowing purposes. Hence no plots were laid down with seed steeped in formaldehyde solution.

Formaldehyde Solution: Spraying.—In order to avoid, if possible, the disadvantages of steeping, attempts were made to disinfect flax seeds by spraying. Lots of half a pound each of affected seed, after having been spread out in thin layers, were sprayed by means of an atomiser with 20 c.c. of diluted formaldehyde solutions of varying concentrations. This amount of liquid was not sufficient to cause the seed to become mucilaginous. After atomising, the seed was dried for twenty-four hours at 40°C. It was then tested for germination, and the viability of the conidia present on the seed was also tested. The results are summarized in the following table:—

Strength of Formaldehyde Solution.	Germination of	
	Seed.	Conidia.
Untreated.	94 per cent.	Abundant.
0·09 per cent.	92 ,,	,,
0·37 ,,	94 ,,	Sparse.
0·48 ,,	85 ,,	Very sparse.
0·59 ,,	73 ,,	Nil.
0·74 ,,	63 ,,	,,
1·49 ,,	51 ,,	,,
3·74 ,,	22 ,,	,,
37·40 ,,	2 ,,	,,

It will be seen that a concentration of 0·59 per cent. of formaldehyde was required in order to kill all the conidia, but the germination of the seed so treated was considerably reduced. Higher concentrations had a more pronouncedly deleterious effect on the germination of the seed, while lower ones did not entirely kill the conidia.

Seed treated with 0·37 per cent., 0·48 per cent., and 0·59 per cent. solutions respectively was sown in experimental plots, and untreated seed was sown in a control plot alongside of them. The disease became evident in the control plot and in the 0·37 per cent. plot as soon as the crop had brairded, and these plots showed "stem-break" and "browning" well developed at the time of pulling. The disease appeared much later in the plot sown with seed treated with the 0·48 per cent. solution, and at pulling time "browning" and "stem-break" were less evident in this plot than in the two just referred to.

The plants derived from seed treated with 0·59 per cent. solution showed no signs of the disease, and they remained quite free from it through the season.

In the three cases where treated seed was sown, the crop was considerably thinner than that derived from untreated seed, and thinner than might have been expected, judging from the results of the germination tests of the treated seed. It is clear that by spraying affected seed with a limited quantity of a 0·59 per cent. solution of formaldehyde the disease can be suppressed. But the quantity of seed sown per unit area would have to be increased considerably to ensure a crop of sufficient thickness, if this mode of control were adopted; and this might not be a profitable undertaking.

Formaldehyde Gas.—Affected seeds were exposed to formaldehyde gas when driven off by the action of potassium permanganate on a concentrated solution, as well as when allowed to evaporate naturally from the solution into a confined space of air. The results so far obtained, however, have not been satisfactory, for, although the conidia are killed, the vitality of the seed is much too seriously affected. Seed treated by the latter method was sown, and the resulting plants arose and remained free from the disease, but they were very few in number. It is hoped to carry out further trials with this gas, paying special attention to time of exposure of the seed and temperature of the gas.

Heat.—A good deal of experimental work was done in connexion with the effect of heat on flax seed. Thus, infected seeds were subjected to a temperature of 50°C. for periods varying from six to one hundred and ninety-two hours. The effect on the fungus was *nil*, the conidia germinating just as vigorously after heating as before, while the general effect on the seed was to improve its percentage of germination.

The conidia on affected seeds were also found to be unharmed by exposure to a temperature of 70°C. for periods of from three to seventy-two hours. Tests of the seed made just after treatment showed a considerable depression in the percentage of germination, but after a period of rest, at room

temperature, this percentage regained its normal amount. Similar results followed heating to 75° C. and 80° C. for periods up to 120 and 168 hours respectively.

Affected seeds were ultimately heated at 96·5°C. for periods varying from three to thirty hours. The conidia survived a twenty-four-hour period of treatment at this temperature in a viable condition, but not one of thirty hours. Seed heated to this temperature for so long a period, however, was found to be permanently injured. It is true that the percentage of germination improved somewhat after a period of rest at room temperature, but it never subsequently reached that of the unheated seed, the highest being 54 per cent., while that of the unheated seed was 94 per cent.

Seed treated in this manner was sown in a plot, but, as was to be expected, the resulting stand of plants was very thin. Moreover, they showed a lack of vigour from the start, and they remained much stunted and dwarfed promising a very poor yield of fibre. Nevertheless, the plants remained entirely free from "browning" and "stem-break" throughout the season.

From what has been said it may be inferred that the solution of the problem of disinfecting infected flax seed without injuring the seed itself is not an easy one.[1] Still, it is not to be regarded as hopeless, but rather as one requiring more intensive investigation; and further experimental work on the matter is already in progress.

No special study has yet been made regarding the question of the possible existence of strains or varieties of flax resistant or immune to the disease. But it has been observed, contrary to what was thought in some quarters, that both blue- and white-flowered varieties are liable to attack.

VIII.—Summary.

The present paper is concerned with a serious disease of flax which occurs in the field in two forms, viz.—(1) "browning," a turning brown of the plants, which occurs rapidly, especially under moist weather conditions, at or before pulling time; and (2) "stem-break," the fracture or partial fracture of the stems of a certain proportion of affected plants, usually in the region of the first node, and thus at or near soil-level.

One and the same fungus was found constantly associated with the disease in both of its forms.

[1] In the trials made so far, death of the conidia present has been taken as the criterion of disinfection of the seed. But it must be remembered that mycelium is also present in the tissues of the seed-coat; and this may possibly remain alive and capable of developing new conidia when those already present are killed.

This fungus was isolated, studied in pure culture, and was proved by means of controlled inoculation experiments to be the cause of both "browning" and "stem-break."

Not having previously been recorded, the fungus is described under the name *Polyspora lini* n. gen. et. sp.

Diseased plants produce infected seed, and it was proved that transmission of the disease occurs through the sowing of such seed. It is probable that this is the chief, if not the only, means by which the disease is perpetuated.

The fungus has been found on flax seed definitely known to have been produced in England and Scotland, as well as on seed purporting to have come from a crop grown in Belgium. It has also been found on flax grown in Ireland from seed believed to have been derived from crops grown in England, Scotland, Holland, Russia, Canada, and Japan, and the disease has been reported as being present on flax in British East Africa. It would appear, therefore, that the disease must be present practically wherever flax is cultivated. It is possible, however, that, owing to different climatic conditions, it may not be so serious in certain of these countries as it is in Ireland.

Seed for sowing purposes should not be saved from a crop suffering from this disease. Seed disinfection trials on a small scale have shown that atomising with a dilute aqueous solution of formaldehyde is one of the most promising means of control when healthy seed is not available for sowing.

Both white-flowered and blue-flowered varieties of flax have been found susceptible to the disease.

EXPLANATION OF PLATES.

PLATE VIII.

FIG.

1. Naturally affected flax seedlings, seen from above, showing dark diseased areas on the cotyledons, attacked by *Polyspora lini*, which represent the earliest stage of "browning." (Slightly enlarged.)

2. Portions of stems of mature flax plants, showing various stages of the "browning" disease. That on the extreme left shows spotting of the leaves only. Death of the leaves and infection of the stems is evident on the remaining four. (× $\frac{1}{3}$.)

3. Bottom row: healthy flax fruits. The upper two rows show affected fruits in various stages of attack. (Natural size.)

4. Transverse section through a mature flax stem affected with "browning." The dark diseased cortical and epidermal tissues extend nearly, but not quite, around the stem. (× 30.)

5. Five naturally affected young flax plants, showing stages in the development of "stem-break." The two on the extreme left show cotyledonary infection only; the two on the extreme right have their cotyledons destroyed, and infection of the stems at the node has occurred, with incipient break. The central one shows an intermediate stage. (Slightly reduced.)

PLATE IX.

1. "Stem-break" produced by placing conidia of *P. lini* from a pure culture on the cotyledons of seedlings. In all cases the stems became attacked, and fracture of the stem, with subsequent upward bending, is well marked in four cases. (Slightly reduced.)

2. Stellate form of growth of *P. lini* on wort gelatine from a single conidium; culture five days old. The masses of conidia are seen along the radiating hyphae, which are, however, invisible in the photograph. (× 30.)

3. Conidia formation in *P. lini*, from a five-day old pure culture on a wort gelatine film. (× 270.)

Plate X.

All figures × 510 diameters.

FIG.

1. Conidia of *Polyspora lini* from a naturally affected flax leaf.
2. Conidia germinating on beef-extract gelatine; twenty-four hours old culture.
3. Same as 2; forty-eight hours old culture, new conidia being produced.
4. Same as 2 and 3; seventy-two hours old culture.
5. Vertical section through portion of an affected flax leaf, showing one conidiophore of *P. lini* protruding through a stoma, and producing conidia.
6. Portion of mycelium from a fifty-four-day old culture of *P. lini* on oat-extract agar, showing the production of conidia on minute lateral outgrowths.
7. Portion of section through the seed-coat of an affected flax seed, showing hyphae of *P. lini* in the outer layers of cells, and two conidiophores, which have produced conidia on the surface.
8. Conidia of *P. lini* produced within an empty cell of a hypha; from a fifty-four-day old culture on oat-extract agar.

XXIII.

A NEW PRINCIPLE IN BLOWPIPE CONSTRUCTION.

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Read April 26. Published August 19, 1921.

The design of a blowpipe which will be suitable for glass-blowing either in chemical and physical laboratories or on a commercial scale, is a matter which does not seem to have received the amount of attention it deserves. Though there are many forms of quick-change blowpipes in existence which are supposed to operate with great ease, none of them that have come to the writer's notice will operate really satisfactorily over the wide range of work usually required of such an instrument, even when used with a foot-bellows, and when used with a power-blower they become still less efficient.

This seems to arise from the fact, which instrument-makers appear to have overlooked, that a blowpipe which is to be used with a power-blower must be differently designed from one to be used with a foot-bellows, if it is to give satisfactory service. This oversight has led to an erroneous conclusion which is frequently seen stated, even in quite modern text-books on glass-blowing—namely, that the foot-bellows is preferable as a source of air to a power-blower, owing to the greater control it gives the worker over the flame. It is now generally conceded, however, that the power-blower enables a greater amount of work to be done with less practice, even with an ill-designed blowpipe, owing to the fact that the attention of the worker is not diverted by the working of a bellows.

It has long been the opinion of the writer that the inconvenience which arises in the use of existing types of blowpipes is due very largely to the fundamentally wrong design and construction of these instruments, and can only be remedied by bringing the design more into line with theoretical considerations.

Although the problem has been worked at from this point of view, the chief practical considerations have also been kept in view, namely, that (1) the blowpipe should give a great range of flames with the minimum of

attention from the worker; (2) it should operate satisfactorily with air at a constant pressure; and (3) it should be compact in size.

General considerations lead to the conclusion that in order to obtain flames of widely different sizes and the same character, with air at a constant low pressure, the air-jet must be variable in bore. For each size of flame there is a particular size of air-jet, which will give a well-proportioned blowpipe flame with a given air-pressure; hence one air-jet cannot be made to serve flames of different sizes satisfactorily. With blowpipes operating with a fixed air-jet and foot-bellows, this defect is partially overcome by varying the air-pressure with the size of the flame, thus supplying somewhat more air to the larger flames by more vigorous working of the bellows. Even this compromise is not quite successful, since, if the jet is of the correct bore for the smaller flames, it cannot supply enough air for the larger ones even with greatly increased pressure. When such a blowpipe is used with a power-blower, however, this difficulty becomes still greater, for it becomes necessary either to vary the speed of the motor for each change in size of flame, or to keep the motor running at the speed required for the largest flame, and vary the air-pressure by means of a tap.

It was, therefore, taken as a fundamental condition that for a blowpipe intended to work with air at a constant pressure, the air-jet must be variable in bore. The next consideration was to find a way in which this could be achieved without much difficulty. Two methods naturally suggest themselves—(1) to arrange for the actual variation of the air-jet bore by mechanical means; and (2) to provide a different-sized jet for each size of flame, and to insert each jet when using the corresponding flame.

The first method is mechanically possible, but it would involve the use of tools and processes which were not at the disposal of the writer, so that attention was mainly directed to the second method. This method is used in an existing type of blowpipe, which is usually called the "glass-blower's pattern" blowpipe, by providing a very wide air-jet, into which short pieces of capillary glass tubing can, with a certain expenditure of time and patience, be wedged with cork or paper to provide the different-sized jets.

However, it is hardly possible that this device was ever intended to be availed of in the course of work, but was rather intended to alter the blowpipe when changing from one type of work to another, since the time required to change the jet would be more than sufficient to allow the glass to cool in the course of any one operation.

The problem then narrowed down to finding a method by which the substitution of different-sized air-jets could be made extremely rapidly, with slight effort on the part of the operator, as the size of the flame varied. If

the operation of changing the jets could be "codified" so as to be performed mechanically, obviously the effect would be quite as good as an air-jet of variable bore.

Consideration of the possible ways in which this might be accomplished directed attention to the tubular shape, which is invariably seen in all types of blowpipe, and it was quickly realized that this shape, if retained, would render the design of a compact blowpipe, such as was contemplated, extremely difficult, if not impossible.

At first it was thought that the persistence of this shape in all types of blowpipe was due to the fact that it was necessary for the proper functioning of the blowpipe; but a few experiments soon showed that the tubular shape had little or no effect on the shape of the flame, and was by no means an essential feature of the blowpipe. The persistence of this shape, which only tends to make the blowpipes more unwieldy and awkward than they otherwise would be, appears to be due to the fact that the original blowpipe was made in this form, and later designers have merely modified it, without attempting any radical departure.

It was therefore decided to abandon this tubular form altogether in the design of the blowpipe to be described in this paper, and to keep the gas chamber as compact as possible, as it had been shown that this could be done without sacrificing efficiency in any way. By doing so it was found comparatively easy to design a blowpipe fulfilling the conditions which were deemed essential.

The objects aimed at in the design to be described are:—

1. To enable the change from one flame to another to be made with the minimum of attention.

2. To vary the air-supply by means of jets of different bore, the air-pressure being constant.

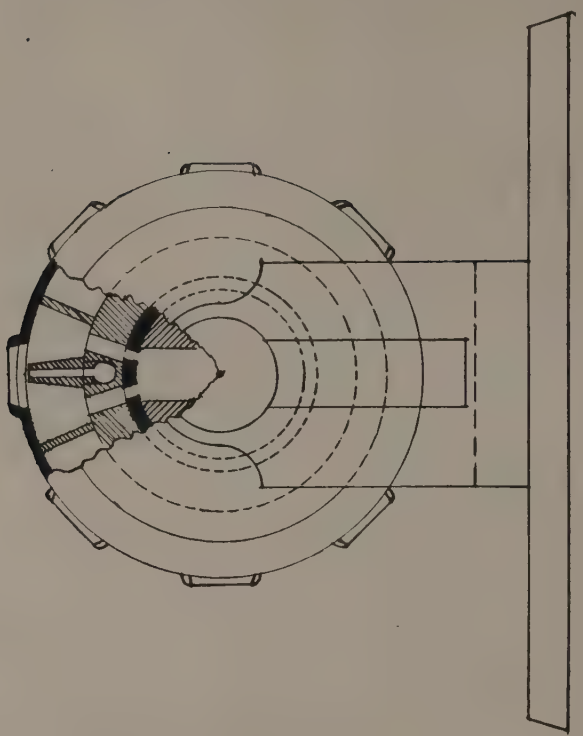
3. To vary the gas supply simultaneously with the air.

4. To enable any air-jet to be used with any given size of flame, in order to vary the character of the flame.

5. To make provision for a flat flame.

6. To keep the whole as compact as possible.

The principle on which the instrument is based consists in placing a number of small gas chambers (each provided with an air-jet of different bore) round the periphery of a cylinder, mounted on a horizontal axis, through which gas and air are supplied in such a way that any size of flame can be brought into operation by mere rotation of the cylinder.



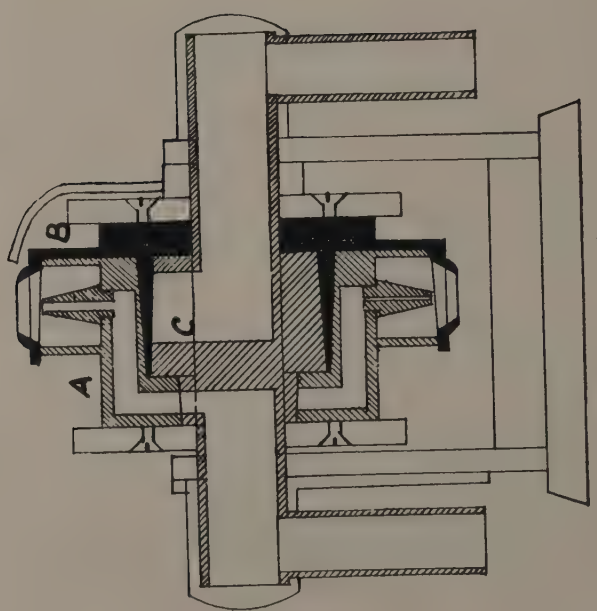


FIG. 1.

A sectional drawing of the experimental blowpipe which has been made to embody and test this principle is given in fig. 1. The instrument consists of two moving and one fixed part. The member which controls the air-supply is marked A in the drawing, and consists of a cylindrical block of brass, in which are drilled eight radial passages for the air, which communicate with eight air-jets. These air-jets are spaced equidistantly round the circumference of a deep recess formed in the brass cylinder. This recess is divided into eight equal spaces by eight brass partitions, thus forming the spaces for the gas to surround the air-jets. The gas has access to these spaces through a number of radial holes drilled in the brass cylinder, as shown in the sectional side elevation in fig. 1. The outer circumference of A is turned taper, and ground to fit the corresponding part of B, and it also has a deep conical hole bored in it for more than two-thirds of its depth, which is also ground to fit the corresponding portion of B.

The part B consists of a thin brass disc, carrying on its periphery a thin hollow cylinder, which is ground taper to fit the outer circumference of A, and is provided with eight apertures through which the flame is projected. This disc also carries a projecting portion near the centre, which has two conical surfaces, the outer fitting the conical hole in A, the inner fitting the corresponding tapered portion of C.

In this double conical portion of B there are drilled a number of equally spaced holes, which, by their size and number, vary the amount of gas admitted to the blowpipe. Two of these holes are indicated in the part-sectional side elevation (fig. 1).

The fixed portion C also consists of two oppositely tapered cones, the right hand one being tapered to fit B, and having a passage for the gas formed in it at one place, the other being tapered to fit the inner circumference of A, and provided with a passage for the air so placed as to correspond with that for the gas.

The portions A and B are free to revolve, both with respect to each other and with respect to C, but a small spring catch secures A in any desired relative position with respect to B, the two then revolving as a solid mass round C. In this way any desired air-jet on A can be brought opposite any gas-jet on B at will.

When A and B are locked together in any position, the revolution of the whole round C gives eight flames of different sizes between about one inch and twelve inches in length, and each of these flames is formed by an air-jet of just the right bore for its particular requirements; thus the bore of the largest air-jet is about ·125 inch, and that of the smallest about ·015 inch.

A by-pass is provided, which is opposite the passages bored in C, and

serves to ignite each flame as it comes into line with it by the revolution of the blowpipe. The parts A and B are provided with fibre discs with milled edges, to enable them to be easily turned when hot. The tube on which the whole is supported is held in simple trunnion bearings, and provided with a quadrant, which allows of the flame being tilted to any desired position. The gas and air are led in through short tubes which are placed at 180° to the position taken up by the flame. The whole is mounted on a simple stand of heavy metal, provided with holes for screwing to the bench.

This design of blowpipe, based as it is on theoretical considerations, naturally admits of modification and adaptation to meet particular purposes; and it will not be out of place to indicate some of the chief advantages inherent in this form.

Owing to the provision of a number of different air-jets for the different flames, the range of this blowpipe is very great, when we consider the small size of the actual working parts. The series of flames may start with a small flame about the size of that produced by a fine mouth blowpipe, and extend to a size which is limited only by the size of the blowpipe and the diameter of the gas-pipe, and yet each flame will be perfectly formed, since it is supplied with a jet of just the right size for its particular requirements. This result is obtained with gas at about four inches of water, and on air-pressure of about twelve inches of water. As well as this succession of round flames, one or more flat blowpipe flames may be produced by suitably altering the gas and air orifices.

The change from any one flame to any other is instantaneously made by mere rotation of the blowpipe on its axis, without any adjustment of taps, with the assurance that a perfect flame of the size desired will be produced in all cases. The character of the series of flames can be altered by the relative rotation of the two discs, thus producing a series of softer or harder flames according to the direction of rotation.

A further advantage is to be found in the fact that once the ratio of gas to air in any one flame is fixed (by inserting a suitable jet) the flame becomes standardized, and can be exactly reproduced at any time by using the same gas- and air-pressure. Consequently, if on one occasion any particular flame is found suitable for any operation, either in glass-blowing or general laboratory work, by noting the number of the air-jet, this flame can be reproduced easily at any future time with great precision, thus leading to the saving of time and the production of uniform results.

The principle also admits of an important development from the manufacturing point of view, inasmuch as it is possible by inserting the correct jets to arrange so that the blowpipe will give a series of flames adapted to

some particular series of operations which has to be carried out repeatedly. Thus, in making any piece of glass apparatus in quantity the same series of operations occur regularly, and the blowpipe could be arranged to give in succession the flames most suited to the different operations.

In making a small gas wash-bottle of glass, for instance, the following series of operations might occur (fig. 2):—

1.	Draw out the glass tubing and seal off .	large flame.
2.	Round the end of large tube . . .	medium „
3.	Blow out end of large tube . . .	very small „
4.	Seal in the small tube	small „
5.	Blow bulb on large tube and seal off . .	large „
6.	Blow out pip for side tube . . .	very small „
7.	Seal on side tube	small „
8.	Bend side tube to shape . . .	large „

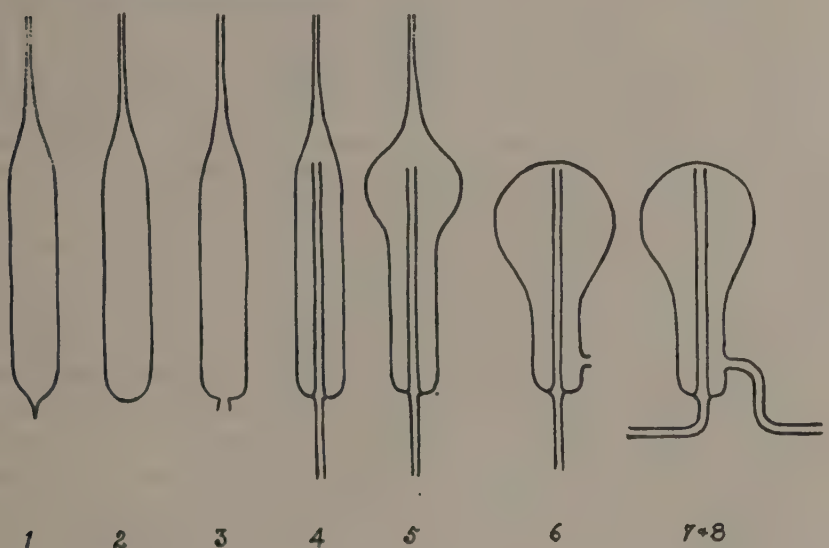


FIG. 2.

If this series of operations had to be carried out repeatedly, it would be a great saving of time to have the blowpipe so arranged that the particular flame best suited to each operation should come into operation in succession at the moment it was required.

Such an arrangement of flames could readily be obtained with the type of blowpipe described.

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XXIV.

UNCHARGED NUCLEI PRODUCED IN MOIST AIR BY ULTRA-VIOLET LIGHT AND OTHER SOURCES.

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AND
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[Read May 24. Published August 19, 1921.]

I.—Nuclei produced by Ultra-violet Light.

Ultra-violet light acting on moist air or oxygen produces in the gas small particles or nuclei which are not electrically charged. C. T. R. Wilson, in a paper on condensation nuclei (Proc. Roy. Soc., London, **64**, p. 127), describes some experiments with these condensation nuclei, which he detected and studied by means of his expansion apparatus. When the radiation was weak, he found that the nuclei produced required as great a degree of supersaturation to cause water to condense upon them as the ions produced by X-rays, uranium, &c. With stronger radiation, however, the nuclei grew in size, requiring a smaller degree of supersaturation to produce condensation. Under favourable conditions they even became visible.

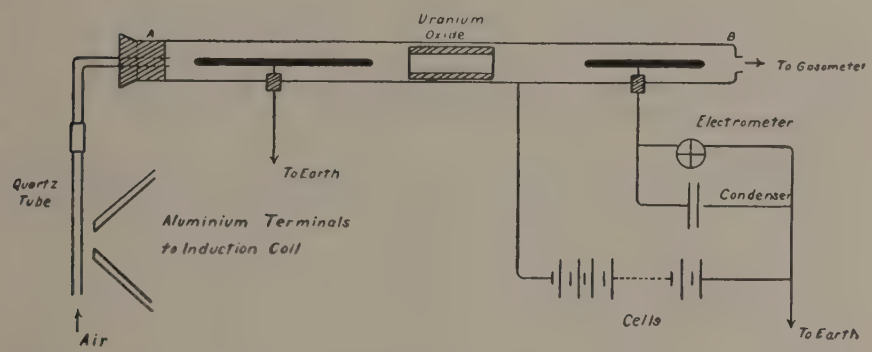
In our experiments the nuclei were detected electrically. Wilson's method of supersaturation gave but a rough idea of the size of the uncharged nuclei, and it could not be applied to dry air. In the following paper an account is given of an attempt to examine the nuclei more closely by an electrical method. Some experiments were also performed on other types of uncharged nuclei.

When uncharged nuclei pass through air ionized by uranium, X-rays, &c., they are electrified by the small ions, and thus large ions are formed, of smaller mobility, under an electric field. These large ions recombine less rapidly than the small ions; and hence, if time is allowed for the ions to recombine, the presence of nuclei originally uncharged causes an increase in the current measured.

P. Lenard and C. Ramsauer have also studied in great detail the nuclei produced by ultra-violet light. Using light of such short wave-length that it was able at once to ionize the air and to produce the nuclei, they obtained large ions, which they attributed to the combination of nuclei and ions pro-

duced separately. They quote an interesting experiment by Becker. Air was filtered and exposed to Röntgen rays, and it was found that only the ordinary small ions were produced in it. If the air, however, before being exposed to the Röntgen rays were first exposed to ultra-violet light, numerous large ions were found to be present. This leads to the conclusion that the large ions are formed by the collision of the small ions with the nuclei. It may be pointed out here, however, that the experiment is not quite conclusive, as the large ions might be produced by the photo-electric effect of the Röntgen rays on the uncharged nuclei themselves. It is shown later on in this paper that ultra-violet light, of wave-length too long to ionize the air, can cause the emission of electrons from uncharged nuclei driven off glass tubes by heating, and it might be argued that a similar process took place in Becker's experiment. Our experiments, however, in

— APPARATUS —



which ionization by the α-rays of uranium oxide took the place of the Röntgen rays of Becker's experiment, and in which positive and negative ions behaved similarly, prove conclusively that nuclei and small ions are separately produced, and by their combination with each other form the large ions.

The diagram gives a plan of our apparatus. Air was drawn by means of a gasometer through a cotton-wool plug, and then over a water surface or through drying tubes if desired. It then passed through a long quartz tube into an ionization tube containing two terminals. The first, T_1, was 50 cms. long, and generally connected to earth. The second, T_2, was 12 cms. long, and connected to a quadrant electrometer with a capacity in parallel. Between the two terminals was a glass tube which just fitted into the ionization tube, and which was coated on the inside with uranium oxide.

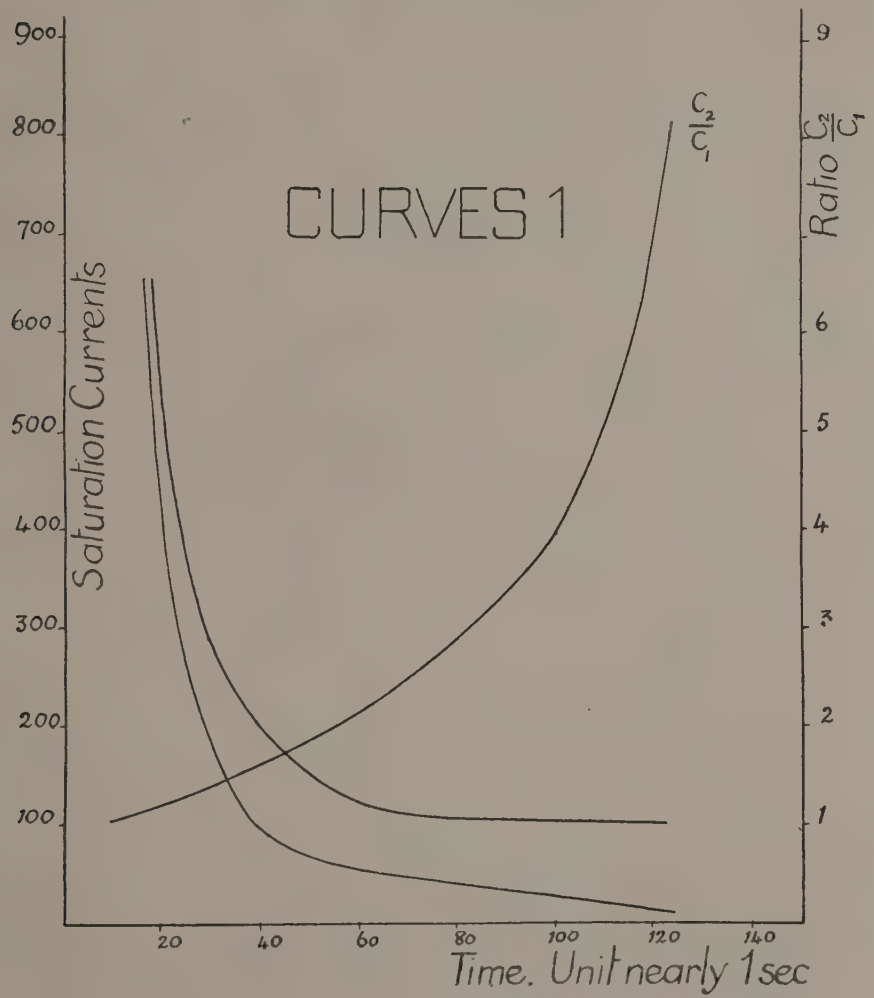
The quartz tube passed through a box lined with tinfoil and earthed. Inside the box, and opposite the quartz tube, were aluminium terminals connected to the secondary of an induction coil. The shape of the terminals enabled them to be placed, if necessary, right up against the quartz tube. A motor-driven mercury interrupter was used with the induction coil. The quartz tube was about half a millimetre thick. In most of the experiments the light also traversed 2 or 3 mms. of air.

The ultra-violet light of the spark was found, in a number of careful experiments, not to ionize the air; but as a precaution the long terminal, T_1, was used to remove to earth any ions contained in the air before its passage over the uranium oxide. The outer cylinder of the whole ionization tube AB was kept at any desired voltage by means of cells. When the air was passing through the apparatus, the ions produced by the uranium oxide took on an average about twenty or thirty seconds to reach the terminal T_2. A great number were consequently lost by recombination. The current due to the uranium oxide itself we shall call C_1. When C_1 had been measured, the spark was started. The air now passing over the uranium oxide contained the nuclei produced by the ultra-violet light. Some of these nuclei attached themselves to small ions. In this way large ions were produced which recombined less rapidly than the small ones. More ions consequently reached the terminal T_2 than when the nuclei were absent, and consequently the observed current C_2, when the spark was on, was greater than the current C_1. The current C_2 will evidently increase with the number and size of the nuclei present in the air, and thus the ratio $\frac{C_2}{C_1}$ gives an idea of the number of nuclei present. An idea as to the relative sizes of the nuclei and small ions can be got from a knowledge of their mobilities under an electric field. This mobility can be deduced from the saturation voltages in a current-voltage curve when the dimensions of the terminal and ionization tube and the quantity of air passing per second through the apparatus are known.

In our experiments, the quantity of air flowing per minute varied approximately from 1000 ccs. to 6000 ccs. The ratio $\frac{C_2}{C_1}$ varied from about 8 for slow blasts of air to about 1·2 for quick blasts. The value of the ratio $\frac{C_2}{C_1}$ is increased for slow velocities of the air through the apparatus, not only on account of the larger time allowed for recombination, but because the time of exposure of the air to the light is also increased, so that more and larger condensation nuclei are produced. For the quickest air-blasts used the nuclei had a mobility of about ·3 cms. per sec. per volt/cm.

For very slow blasts large ions were found with a mobility as low as ·00024. The slower the air-blast the smaller was the mobility.

Curves 1 represent the manner in which C_2 and C_1 decay with the time taken by the air to traverse the apparatus. The currents are, as throughout, in arbitrary units. The manner in which $\frac{C_2}{C_1}$ changes with time is also



shown. C_1, of course, is the current of small ions due to the uranium by itself when no spark is playing. C_2 is the current of large ions due to the charging by the uranium ions of the nuclei produced by the ultra-violet light. The air in all these experiments passed over a water surface. For the

quickest flow of air (7,800 ccs. per min.) the value $\frac{C_2}{C_1} = 1{\cdot}06$; for the slowest (812 ccs. per min.), $\frac{C_2}{C_1} = 8{\cdot}1$.

Mobilities met with in these Experiments.

The mobility of the charged condensation nuclei varied with the rate of flow of the air through the apparatus. The slower the rate of flow the larger were the ions. The mobilities obtained varied from ·33 to ·00024 cms. per sec. under a field of 1 volt per cm. We obtained ions of mobilities ·33, ·18, ·10, ·02, ·008, ·0044, ·0034, ·0025, and ·00024. The numbers were deduced from the saturation voltages in current-voltage curves, and do not claim a high degree of accuracy. The positive and negative ions under similar conditions have the same mobilities.

As an example of the manner in which the mobility varied with the rate of flow of the air, the following numbers will be quoted. During the experiment the intensity of the light, &c., remained constant, only the rate of flow of the air varying:—

Quantity of Air per min.	Mobility.
3600 ccs.	·18
2050 ,,	·008
1120 ,,	·0034

Effect of Varying the Intensity of the Light.

C. T. R. Wilson observed that the nuclei produced by ultra-violet light increased in size as the intensity of the light increased. Several experiments were performed by us showing that by diminishing the distance of the spark from the quartz tubes, and thus increasing the light intensity, the nuclei grew in size and became more numerous.

In these experiments, keeping the current of air constant, and a voltage on the ionization tube sufficient to "saturate" the largest ions produced, the distance of the spark was varied. One of the quartz tubes employed gave an appreciable effect up to 15 mms. distance; with the second the effect stopped at 5 mm. Thus in one experiment, calling the current due to the uranium alone, $C_1 = 100$, the following values were obtained for C_2 with

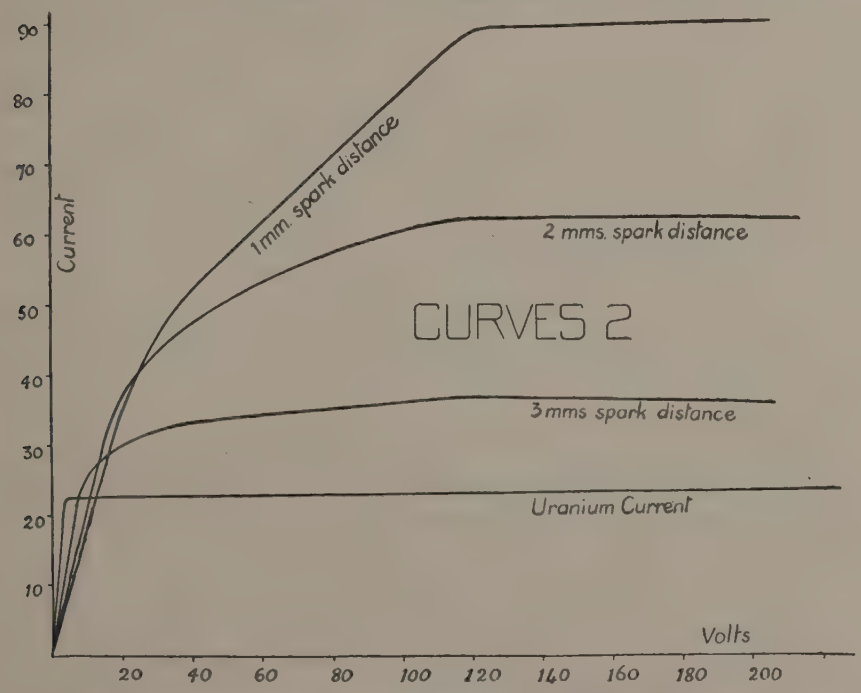
the spark at different distances, the quantity of air per minute being 2,000 ccs. :—

Distance of Spark in mms.	C_2 = current due to charged nuclei.
5	213
10	162
15	117
No Spark.	$C_1 = 100$

In another experiment, using a different quartz tube and a blast of air = 3,000 ccs. per minute, the following numbers were obtained :—

Initial uranium current, $C_1 = 236$.
Final „ „ $C_1 = 238$.

Spark distance in mms. .	= 15	10	5	3	2	1
Current C_2 . . .	= 238	250	250	365	625	910



In the latter experiment a current-voltage curve was obtained for each distance of the spark, and also one when the spark was stopped, giving the current due to the small ions alone. Curves 2 show the relative magnitude of the currents at different voltages. All the curves have the same saturation

point near 120 volts, indicating an ion of mobility about ·009. For the larger spark distances this ion is present in small quantities only. Thus when the spark is 3 mms. from the quartz tube, the largest ion is about 16 per cent. of the total; at 2 mms. 40 per cent.: at 1 mm. 61 per cent. The data from which the curves were drawn are given in the following tables :—

TABLE FOR CURVES 2.

Spark distance (mms.)	Volts.	Current.	Distance.	Volts.	Current.
No Spark	200	23·6	2 mms.	200	62·5
,,	40	22·3	,,	120	62·5
,,	20	23	,,	80	58
15 mms.	200	23·8	,,	40	47·6
10 ,,	200	25	,,	20	36·5
5 ,,	200	25	,,	10	20·8
3 mms.	200	36·5			
,,	120	37	1 mm.	200	91
,,	80	35·2	,,	120	89
,,	40	33·3	,,	80	71·4
,,	20	31·3	,,	40	52·1
,,	10	25	,,	20	35·2
			,,	10	18·5

Similar behaviour of (+) *and* (–) *Ions.*

Our experiments showed that no difference existed between positive and negative numbers; or, in other words, that the nuclei attached themselves indifferently to positive or negative small ions.

A current-voltage curve was plotted, using positive and negative voltages alternately. A preliminary test had shown that when the first terminal T_1 was earthed, and the uranium removed, no ions (due to the spark), either positive or negative, reached the second terminal T_2. The following table gives the numbers observed :—

	Voltage.	Current.
Uranium alone, . .	+ 120	116 (C_1)
	– 120	133 ,,
Uranium and Spark, .	+ 120	833 (C_2)
	– 120	806 ,,

Ionization Curve (Uranium and Spark).

Volts positive.	Current.	Volts negative.	Current.
10	238	10	246
20	400	20	382
30	467	30	500
40	575	40	543
60	625	60	667
80	735	80	676

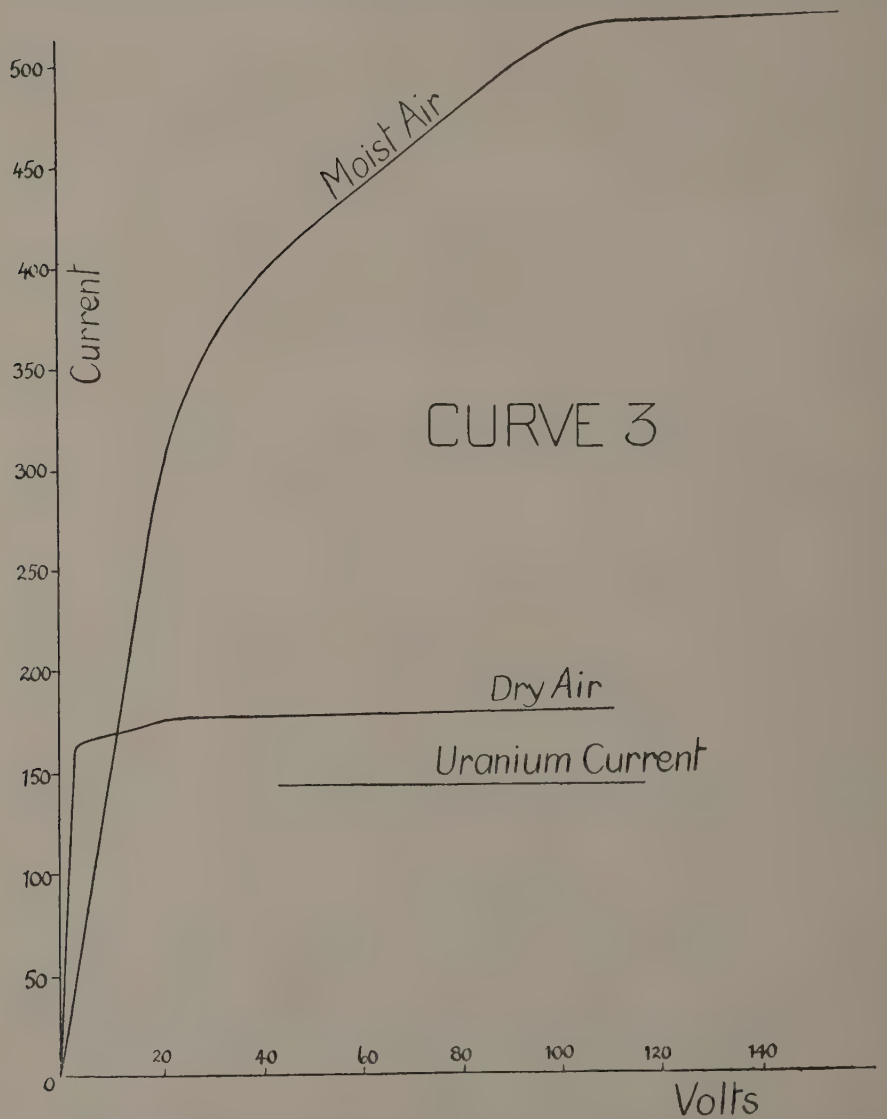
It will be evident from the tables above that positive and negative numbers are materially the same, indicating the equal behaviour of positive and negative ions.

Results similar to the preceding were obtained by Lenard and Ramsauer (*loc. cit.*). In their experiments the large ions were produced directly by Schumann—violet rays, which at once ionized the air and produced the nuclei. They found that the ions increased in size with the intensity of the light, the mobilities being measured, as in our experiments, by plotting a current-voltage curve. Increasing the time of exposure of the air to the light by diminishing the velocity of the air past the source of light also increased the size of the ions, as in our experiments. Lenard and Ramsauer give curves showing the decrease of the currents by recombination (cf. Curves 1); the rate of decrease being slower, the more vaporous impurities are contained in the air. Finally, the mobilities of the positive and negative ions were shown by them to be the same. The mobilities were deduced from the saturation points in a current-voltage curve.

Effect of Drying the Air.

The number and size of the nuclei depend upon the amount of moisture in the air. Thus the effect almost disappears in air dried by calcium chloride, but even on drying with phosphorus pentoxide a small number of nuclei seem always to be produced, as shown by a slight increase in the uranium current on switching on the spark. Curves 3 show the relative effects in moist and dry air. As will be seen, the current C_2, consisting of condensation nuclei charged by the uranium ions, is in the case of dry air only slightly greater than C_1, the current due to uranium alone, while in the case of moist air C_2 is $3\frac{1}{2}$ times as great as C_1. The saturation point in moist air is 40 volts, indicating an ion of mobility ·014. Furthermore, these ions

of mobility ·014 are nearly 50 per cent. of those formed in moist air. In dry air the largest ion is saturated at 8 volts, indicating an ion of mobility ·07, which, as is evident from the curve, forms only about 7 per cent. of the total.



In all probability rigorous drying would remove the effect altogether, but in the experiments tried by us a small number of nuclei was always formed in dry air. The effect in ordinary air depends, of course, upon the humidity of the atmosphere, but is less than the effect in air passed over a water

surface. Thus in one experiment the uranium current was increased by the spark five-fold in ordinary air, and seven-fold in air passed over a water surface. The air was filtered in both cases as usual.

Nature and Origin of the Nuclei.

It has been pointed out that the slower the moist air passed the spark the greater were the nuclei produced. If moist air at rest be exposed to strong ultra-violet light for about thirty seconds, the nuclei grow so large that they become visible, and the path of a strong beam of light through the vessel reveals a dense blue cloud, the individual particles of which can be seen in a microscope with a dark-field arrangement. Those particles are evidently drops of H_2O, and thus the nuclei which we dealt with in the preceding experiments were in all probability minute drops of water formed by the light, and growing in size with the intensity of the light and the time of exposure.

C. T. R. Wilson first attributed the formation of the nuclei to the production of some substance such as H_2O_2, which when dissolved in water lowered the equilibrium vapour-pressure, thus causing drops to be stable and grow which would otherwise evaporate. Other experimenters have proposed oxides of nitrogen as the parent substance of the nuclei.

In our experiments the difference between the formation of nuclei in dry and moist air has been shown to be very remarkable. The number and size of the nuclei were found to depend upon the quantity of water-vapour present in the air. Experimenters who used Wilson's expansion apparatus to detect the nuclei have stated that carefully dried and moist air behave similarly as to the production of nuclei (St. Sachs. Ann. der Physik, **34**, 1911, and Saltmarch, Proc. Phys. Soc., London, **27**, 1915). In view of our experiments quoted above, this must mean either that the expansion apparatus is so very sensitive a method of detecting nuclei that those formed on the slight traces of moisture left in the air after careful drying cause the formation of an appreciable cloud in the cloud-chamber, but yet would not be numerous enough to affect appreciably the value of the currents in our experiments, or else that the hygroscopic parent substance (e.g., oxides of nitrogen) is formed in dry air, and forms the large nucleus when admitted into the saturated atmosphere of the cloud-chamber. On the latter supposition, if dried air is exposed to the ultra-violet light in our experiments and then passed over a water surface before entering the ionization chamber, large nuclei should be formed and detected by the increase in the current C_2 in the usual way. This experiment was performed, but no nuclei were detected.

A further experiment was performed in which dried air was passed over a

water surface at 0°C. before being exposed to the spark. A small definite quantity of water vapour was thus contained in the air, and a small number of nuclei were formed, $\frac{C_2}{C_1}$ being found to be 1·25. On now including a second water surface at room temp. 11°C. after the quartz tube, it was thought that $\frac{C_2}{C_1}$ would increase, as if, for example, H_2O_2 were formed in the partly moist air more nuclei would be formed in passing over the second water surface. $\frac{C_2}{C_1}$ was found, however, to have the value 1·24 in this case, so that no additional nuclei were formed.

Many physicists have tried to demonstrate the presence of H_2O_2 in the nuclei formed by ultra-violet light in moist air. Vincent (Camb. Phil. Soc. Proc., 1904, **12**) used a sensitive photographic test, but failed to detect any H_2O_2. Miss Saltmarsh (*loc. cit.*) showed that prepared H_2O_2 when introduced into the expansion apparatus caused the production of nuclei. The presence of H_2O_2 in these nuclei could be demonstrated, as, when about 19 expansions were made, the showers falling into a small vessel containing titanic acid, the acid became yellow. On the other hand, 150 expansions on the nuclei produced by ultra-violet light caused no colouration of the acid. Miss Saltmarsh concluded that the part played by H_2O_2 was at most a very small one in the formation of nuclei.

On the other hand, Barkow (Ann. der Physik, **23**, 1907) found that H_2O_2 does not assist cloud-formation unless when decomposed by strong sunlight. When the H_2O_2 is exposed to strong sunlight, numerous nuclei are produced, which persist for long periods. Ultra-violet light is not necessary for this decomposition of H_2O_2, but it was not brought about by light from a strong arc lamp. Barkow tested for hydrogen peroxide in the nuclei produced by ultra-violet light in moist air by drawing them slowly through a solution containing titanic dioxide in concentrated sulphuric acid. This solution turns brown with H_2O_2, but Barkow obtained no colouration even after 150 hours, using in all 134 litres of moist air.

Barkow investigated the effect of nitrogen peroxide on the condensation of water vapour. He passed nitrogen dioxide into the cloud-chamber. Spontaneous condensation immediately took place, and many expansions were required to remove the nuclei. He also ozonized oxygen, and passed it into the expansion apparatus. The characteristic blue cloud was obtained without expansion. This spontaneous condensation with ozonized oxygen, however, Barkow attributed to the presence of small quantities of nitrogen in the oxygen used, oxides of nitrogen being formed.

Pringal (Ann. der Physik, **26**, 1908) verified the effects of oxides of nitrogen upon the condensation of water vapour, and examined Barkow's supposition that the condensation brought about by ozonized oxygen is really due to traces of nitrogen in the oxygen used. The question was a difficult one, owing to the task of obtaining oxygen entirely free from nitrogen; but Pringal set himself to examine the effect of successive purifications of the oxygen used. Oxygen prepared electrolytically was used, and after passing through the ozonizer was passed through a solution of NaOH to remove oxides of nitrogen. The spontaneous condensation still took place, and Pringal was led to believe that the ozone, even when free from oxides of nitrogen, was able to oxidize the traces of nitrogen in the cloud-chamber. Using a series of seven ozonizers, each fitted with a NaOH tube, he partially exhausted the cloud-chamber, and drew the ozonized oxygen, from which the last traces of nitrogen should have been removed, into the cloud-chamber. He repeated this process of exhaustion and replacement by ozonized oxygen, the gas in the cloud-chamber becoming freer and freer from nitrogen. The process was repeated 400 times, and the spontaneous condensation gradually decreased, and finally disappeared. Still a cloud could be formed with expansion, but even the condensation with expansion disappeared gradually. The ozonized oxygen now had no effect on the condensation of the water vapour, and the previously observed condensation was attributed to the oxides of nitrogen. Barkow and Pringal suggest that nuclei formed in moist air by ultra-violet light are similarly due to the production of oxides of nitrogen, and not to hydrogen peroxide.

In this connexion also Miss Saltmarsh examined the production of nuclei in moist nitrogen. When traces of oxygen are present nuclei are readily formed; but when the residual oxygen is removed by sparking for twenty minutes and absorbing the oxides of nitrogen in NaOH, the ultra-violet light produced no nuclei in the pure moist nitrogen.

The case for hydrogen peroxide, being the parent substance, rests upon a test by Bieber (Ann. der Physik, **39**, 1912). He passed moist oxygen exposed to ultra-violet light through a vessel at the low temperature of $-79°$ C. The condensation products were then subjected to the following tests for hydrogen peroxide :—

(1) Potass. iodide and starch, which, in the presence of ferrous sulphate, gives a blue colouration with hydrogen peroxide.

(2) Mixture of potass. ferricyanide and ferric chloride, which gives a blue colouration with H_2O_2.

(3) Dilute solution of titanium dioxide in strong sulphuric acid, which turns brown in the presence of H_2O_2.

All three tests answered for hydrogen peroxide, and the experiment is apparently conclusive. It may be objected, however, that the hydrogen peroxide detected was not present in the nuclei, but simply formed in the air by the ultra-violet light, and not affecting the condensation.

We, accordingly, tried a test in which the nuclei after being charged by uranium were driven by the field on to an electrode covered with moist paper. The paper had been dipped into a mixture of starch solution, potass. iodide, and ferrous sulphate. No blue colouration was observed even after twelve hours. The quantity of H_2O_2 may, however, have been too small to be detected by this method.

The following experiments were performed by Lenard and Ramsauer on the origin of the nuclei which were detected by their effect on the steam-jet. Air was freed from all vaporous impurities by cooling to – 76° C. No nuclei were formed in this air by ultra-violet light. Adding slight traces of water-vapour to the air caused a slight but perceptible effect on the steam-jet. Saturating the pure air with pure water-vapour by passing it over a large water surface produced a marked effect on the steam-jet, but not nearly so strong as that of ordinary moist laboratory air. It is evident, then, that water-vapour is only partially responsible for the production of the nuclei in ordinary air, and that cooling to – 76° C. removes, besides water-vapour, some other effective agent. Slight traces of water-vapour along with vapours from india-rubber gave a strong effect on the steam-jet, while the same amount of water-vapour alone, or of vapour from india-rubber alone, gave only a slight effect. We shall see later in Lenard's experiments upon other vapours, besides water-vapour in air, that ammonia gave many nuclei on exposure to ultra-violet light. It is possible that ammonia is the other effective agent besides water-vapour in ordinary air. Lenard attributes the nucleation in the case of ammonia to the formation of ammonium nitrite, and ammonium nitrate, and, where water-vapour is alone present, to the formation of hydrogen peroxide.

Experiments on Air Saturated with other Vapours.

Experiments were performed on air containing vapours of ethyl alcohol, methyl alcohol, and toluol. The room air, after being carefully dried, was passed over a surface of the liquid before being exposed to the ultra-violet light. C_1 and C_2 were then measured, as in the case of water-vapour, but in all three cases examined C_2 was equal to C_1, so that nuclei similar to those obtained with water-vapour in the air were not formed.

It was noticed in these experiments that if minute traces of either of the three vapours mentioned above were present they prevented the formation

of the nuclei in moist air. In one experiment room air was filtered and passed over a water surface and exposed to ultra-violet light. $\frac{C_2}{C_1}$ was found to be nearly 4. On putting about $\frac{1}{3}$ cc. of alcohol in the water, whose volume was about 1,000 ccs., $\frac{C_2}{C_1}$ dropped to the value unity, so that no nuclei were formed.

The action of the alcohol-vapour in preventing the formation of the nuclei might be in either of two ways. It might prevent the formation of the hygroscopic substance (e.g., H_2O_2) which is the parent of the nucleus, or it might act on the minute water-drops after formation.

To test this point, nuclei were produced in moist air and then passed over an alcohol surface. After passing over the alcohol surface the nuclei could no longer be detected by our methods, the action of the alcohol-vapour being just the same as if it had been contained in the air before exposure to the ultra-violet light.

Air saturated with alcohol-vapour was now exposed to intense ultra-violet light in a closed vessel for some time. A narrow beam of intense arc light traversed the vessel. Its path was at first invisible, but when the spark had acted for thirty seconds or so a thin cloud of large drops was seen in the beam. Using water instead of alcohol, and exposing for the same time, a cloud is also formed, but very different in appearance. It is now blue, the arc light appearing as a solid blue beam through the vessel. The drops in the case of water-vapour are evidently much smaller and more numerous than when alcohol-vapour is used.

The effect of adding traces of alcohol-vapour to the water-vapour in the above experiment is to prevent the formation of the characteristic blue cloud. A thin cloud of large drops is, however, formed similar to that obtained with alcohol-vapour alone. In the light of these results the reason why no nuclei were detected in our experiments by the electrical method is that, although nuclei were formed, they grew so big and were so few that their presence could not be demonstrated by our methods.

Lenard and Ramsauer also investigated other vapours besides water. Alcohol, methyl, ether, benzol, chloroform, atrachinon, and alizarin were found to be ineffective. Benzine was slightly effective in forming nuclei. Ammonia, hydrogen bi-sulphide, carbon-disulphide were effective. In these experiments pure air was passed over a bulb containing water, and another containing the liquid to be examined. In addition, Lenard examined the nuclei formation in moist gases other than air. Oxygen and carbon-dioxide behaved like air; but no nuclei were formed in moist hydrogen. No nuclei were formed in water-vapour alone.

II.—Nuclei driven off Glass Tubes by Heat.

If a glass tube be heated while air is passing through it, uncharged particles are given off which can be charged by passing over uranium oxide. The apparatus previously described was used also in these experiments, the glass tube examined being included between the cotton-wool filter and the quartz tube. A small quantity of charged particles was also given off when a tube was heated.

The tube was generally heated with a luminous gas flame extending over about an inch of the tube. The presence of the nuclei was demonstrated by an increase in the uranium current just as described in the case of the nuclei produced in moist air by ultra-violet light. The emission of the nuclei fell off rapidly, almost ceasing in about ten minutes generally, the current registered falling off to the value of the uranium current alone. The effect reappeared, however, on heating a cool place on the tube.

Thus in one experiment the following numbers were obtained :—

Small ion current (uranium alone) . .	$C_1 = 19{\cdot}8$
Current of nuclei driven by heat from glass and charged by the uranium ions . .	$C_2 = 90{\cdot}9$
After ten minutes' heating . . .	$C_3 = 20{\cdot}8$

The large increase in the current would lead us to expect that the nuclei are of great size. An attempt was then made to plot a current voltage curve. Owing to the fatigue effect it was necessary for each reading to heat a new place of the tube, beginning at that part furthest from the end where the air entered. The charged ions driven off the tube by heat were removed to earth by a second ionization tube as above described, so that the total increase in the current is due to the uncharged nuclei. Saturation occurred at 280 volts, giving a mobility of ·0044. The curve also indicated the presence of a smaller ion of mobility about ·016. The mobility ·0048 was obtained in another experiment.

Another experiment on a different tube did not give saturation at 400 volts—the highest voltage then available. This indicated an ion of mobility less than ·0028. The experiment was repeated, using a longer terminal, and the mobility ·0016 obtained.

The nuclei obtained by heating glass tubes are much more numerous than those produced by ultra-violet light in moist air. In the latter experiments some indications were always found of unattached small ions, whereas the nuclei driven off by heat from glass tubes were so numerous that every small ion attached itself to a nucleus before reaching the terminal.

The tubes which lost on heating the property of emitting uncharged

nuclei regained that property after a certain interval and under certain conditions. The following experiments will be quoted as examples.

A glass tube was strongly heated in a Bunsen flame and "fatigued." It was left open, exposed to the atmosphere for a few days. When again examined it had recovered, the ratio $\frac{C_2}{C_1}$ being almost as great as when the tube was fresh. Heating for fifteen minutes suffices to drive off nearly all the nuclei, the value of $\frac{C_2}{C_1}$ falling off almost to unity.

It might be expected that the effect would be recovered immediately on cooling. This is not the case, nor is the effect recovered if the tube is left in the apparatus overnight closed in. Thus several glass tubes which had been fatigued were found not to recover if left for several days in the apparatus closed in, but recovered fully if left overnight fully exposed to the atmosphere.

The nuclei driven off by heat from glass tubes can also be charged by exposure to ultra-violet light. The apparatus is in this case identical with that described in the previous experiments. The uranium is, of course, excluded. The tube is heated, and the nuclei so obtained are exposed during their passage through the quartz tube to the ultra-violet light of the spark, which is a few mms. distant. A current voltage curve for the positive ions thus obtained gave a mobility of ·0009.

When a tube is heated and fatigued, as above described, the nuclei which can be charged by passing over uranium are recovered in about twenty-four hours when the tube is exposed to the atmosphere. The property of emitting nuclei, which could be charged by ultra-violet light, is not regained so rapidly. Thus a tube which recovered in twenty-four hours the property of emitting nuclei which were charged by the uranium small ions gave off no nuclei which could be charged by ultra-violet light. The latter property, however, it regained in about a week. It may be that the nuclei are not acted upon by ultra-violet light until they reach a certain size, and that in the initial stages of recovery of a glass tube the nuclei given off are small.

The ultra-violet effect is probably due to the photo-electric emissions of electrons from rather big particles. If this is the case, we should expect that many of the electrons would be lost by diffusion to the sides of the vessel before becoming attached to neutral particles. We should thus expect the positive numbers to be greater than the negative.

An experiment was performed with a new glass tube to investigate this point. Positive and negative numbers were taken alternately, the spark running continuously, and the tube heated for each reading at a different

place. The negative numbers were very small compared with the positive the rate of flow of the air being slow (1200 ccs. per min.). The saturation current was ten times as great for positive as for negative numbers. The positive ions were saturated between 320 and 400 volts, giving a mobility between ·0015 and ·0012. The negative ions had a lower saturation point, and were thus smaller than the positive. The numbers obtained are shown in the following table:—

Currents obtained on heating glass tube and charging nuclei by ultra-violet light.

Volts (+)	Current (+)	Volts (−)	Current (−)
80	50	80	13·3
160	100	160	20·4
240	139	240	22·7
320	200	320	21·7
400	227		
440	192		

On the other hand, as might be expected, it was proved in many experiments that when nuclei are driven from a glass tube by heat and passed over uranium, the resulting numbers, positive and negative, are equal, any charged products from the glass tube being previously removed to earth, as in our other experiments.

It was thought possible that passing dry air through the glass tube would remove the matter which gives rise to the condensation nuclei driven off by heat. A glass tube, however, through which dry air had passed for several hours was unchanged, and gave the same effect as before the drying. Whether the air drawn through the glass tube while heated is dry or moist makes no difference in the numbers of nuclei emitted.

In dealing with soft glass tubes a second type of nuclei emission was encountered with stronger heating. If a soft glass tube be heated with a luminous flame, the emission falls off almost to zero in ten or fifteen minutes. If the fatigued part be now heated with a strong Bunsen flame, the emission is very much increased, and remains constant for a long time, no sign of fatigue being observed. In this second case the glass tube glows, and the nuclei are due probably to the decomposition of the glass itself.

III.—Nuclei driven off from Metals by Heat.

A series of experiments were conducted on the nuclei driven off by heat from a platinum wire electrically heated. The platinum wire used was thin, having a resistance per cm. of about ·15 ohms. The air after being filtered and, if necessary, dried, passed through a wide glass tube. Through corks in the ends of the tube thick copper wires were introduced supporting about 5 cms. of the platinum wire. A variable resistance and a sensitive ammeter completed the heating circuit. The wide glass tube containing the platinum wire was heated strongly in a Bunsen flame before being used, to expel all the nuclei from the glass. This precaution was, perhaps, unnecessary owing to the width of the glass tubes, whose diameter was about 6 cms., and which did not become very hot in the experiments.

The remainder of the apparatus was similar to that used in the experiments on nuclei produced by ultra-violet light. The nuclei emitted by the platinum wire were passed over a long terminal connected to earth, then over uranium, and were detected at the short terminal connected to the electrometer. The mobility of the nuclei when charged was deduced from a current-voltage curve as before, and gave an idea of their size.

For simplicity, we shall first deal with the nuclei emitted when the wire has been kept for some time at a dull red heat. The emission then becomes very steady, and seems to be a function of the temperature alone, so that when the velocity of the air through the apparatus remains constant the number and size of the nuclei depend solely upon the current through the wire. For example, if the emission is measured at a given temperature, and the wire be kept then at a much higher temperature even for hours, on decreasing the temperature of the wire to its former value the emission of the nuclei as deducted from the ratio $\frac{C_2}{C_1}$ is found to be unaltered.

It was thought that the nuclei might have been formed by some catalytic action of the platinum upon the moist air, in which the platinum itself was unchanged. Whether the air was moist, however, or carefully dried over calcium chloride and phosphorus pentoxide made not the slightest difference in the numbers. The nuclei were, therefore, due in all probability to the disintegration of the platinum itself.

The size of the nuclei increased with the temperature of the wire. $\frac{C_2}{C_1}$ was measured for currents of 1·2, 1·3, and 1·5 amperes through the wire, the air velocity through the apparatus remaining constant. A current-

voltage curve was plotted for C_2, and the mobilities deduced from the saturation voltages. The results are seen in the following table :—

Current through wire.	$\frac{C_2}{C_1}$	Saturation Voltage (C_2).	Mobility.
1·2	2·5	70	·013
1·3	7·2	120	·008
1·5	> 9·6	> 400	< ·0022

When 1·5 amperes were used saturation did not occur even at 400 volts, the highest voltage obtainable at the time. The numbers, however, show clearly how the size of the nuclei depends upon the temperature of the wire.

In observing the emission from platinum at a given temperature, no falling-off is observed even after hours; but if the same wire is heated from day to day, a gradual falling-off is observed, although as long as the wire is glowing nuclei are always emitted in large numbers.

In addition to the nuclei given off by platinum when glowing, nuclei are also emitted at lower temperatures by fresh wires, but the effect falls off rapidly to zero, and is thus not easily detected. A fresh wire was heated by currents from ·5 ampere up, and the values of C_1 and C_2 taken at each reading of the current. No nuclei were observed at ·5 amp. nor at ·8 amp.; but when the current was 1·0 amp., C_2 was originally four times C_1, but rapidly fell off to a value near C_1, showing that the emission of nuclei fell off to zero. The wire just began to glow at 1·1 amps., and at that low temperature, although the emission never fell off to zero, the wire, when fresh, emitted more nuclei than after it had been heated for some time.

Taking another new piece of wire, the following values of C_1 and C_2 were got for the given currents through the wire. C_1 and C_2 are given in arbitrary units :—

C_1	16
C_2 at 0·5 Amp. . .	16
,, at 0·8 ,, . .	53
Successive readings at 0·8 ,, . .	20
,, ,, at 0·8 ,, . .	15

Thus a large number of nuclei were emitted when 0·8 ampere passed through the wire. Three readings, each taking about one minute, were made

of C_2. The numbers show that the emission fell off to zero in about three minutes.

On increasing the current to 1·0 ampere more nuclei were emitted (although the wire was not glowing). The emission in this case also fell off almost to zero, but took a much longer time than that at ·8 amp. The following values of C_2 and C_1 were got at 1·0 ampere :—

C_1 =	14·5
C_2 (1st Reading)	52·6
,, (2nd ,,)	33·3
,, (3rd ,,)	28·6
,, (4th ,,)	24·4
,, (5th ,,)	23·3
C_1 again	14·5

It will be seen that the rate of emission gradually decreased, but even after about five minutes was still appreciable. On leaving the current at 1·0 amp. for thirty minutes, C_2 was found to be 15·4 and C_1 14·0, so that the emission of nuclei had practically ceased.

On increasing the current through the wire to 1·2 amperes the wire glowed, and C_2 was 37, while C_1 was 15·3. The same values were obtained after twenty minutes' heating.

The initial emission at 0·8 and 1·0 amperes was not regained by the wire even on being laid aside for over three weeks. It is probably due to the emission of occluded substances in the platinum or to surface impurities received, for example, in handling. A fresh platinum wire which was heated in a Bunsen flame before being examined as above did not emit any nuclei at the temperatures corresponding to 0·8 and 1·0 amperes.

With a wire made of ni-chrome, an alloy of nickel and chromium, results were obtained similar to those described for platinum. The wire in this case began to glow at a current of about 3 amps., but nuclei were emitted from the fresh wire at 1·5 amperes. This initial emission rapidly fell off to zero.

The emission at glowing temperatures remained constant for hours, but in using one wire from day to day a marked falling-off is observed. This was clearly shown by the values at 4·0 amps. of $\frac{C_2}{C_1}$, which fell in about a week from the value 5·4 to 2·5, 1·4, 1·25, and finally 1·2.

SUMMARY.

I.

The nuclei produced by ultra-violet light in moist air are detected by an electrical method, and their size indicated by their mobility, when charged in an electrical field. The number and size of the nuclei depend upon the time of exposure to the light, the intensity of the light, and the amount of moisture in the air.

The nuclei are probably small water-drops which owe their formation to the production in the moist air by ultra-violet light of some hygroscopic substance, such as hydrogen peroxide or oxides of nitrogen. An account of work by other experimenters is given, and the evidence, though not conclusive, points to hydrogen peroxide as the parent substance of the nucleus.

Some experiments were performed in air containing, instead of water, the vapours of methyl alcohol, ethyl alcohol, and toluene respectively. No nuclei are found by our method, probably because the nuclei grow so big, and decrease so much in number, that they fail to affect the currents in our apparatus. The presence of very slight traces of either of these vapours in water-vapour prevents the formation of nuclei such as can be detected by our apparatus, and probably for the same reason.

II.

The nuclei which are emitted by a glass tube when heated not too strongly can be detected and measured in a manner similar to that described for the nuclei produced by ultra-violet light. The emission of nuclei when the heating is not too strong lasts only a few minutes, but the tube "recovers" under certain conditions.

The nuclei can be charged by ultra-violet light, probably by the photo-electric discharge of electrons from the rather large particles. The positive ions are found to be much more numerous and of much greater size than the negative. In all probability only particles greater than a certain size are acted on by the light. The electrons then become attached to the smaller particles or the air molecules. The negative ions thus formed are numerically more decreased by diffusion to the walls of the apparatus than the larger positive ions.

The experiments on the recovery of glass tubes fatigued by heat seem to indicate a difference between those nuclei which can be charged by ultra-violet light and those which can be charged by the small ions produced by

uranium. The difference is probably, however, one of size, as in the initial stages of recovery the particles emitted may not be so large as they become after a prolonged period of rest.

Nuclei of a different type are emitted at glowing temperatures. In this case the glass does not become fatigued, and the nuclei are probably produced by the decomposition of the glass itself.

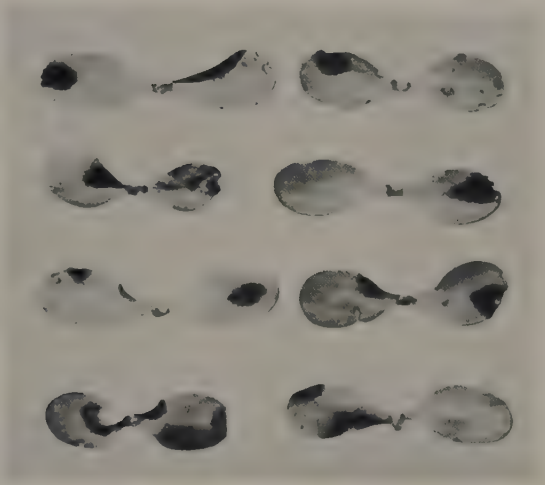
III.

When platinum wires are heated electrically, nuclei are driven off before the wire begins to glow. This emission lasts only a short time, just as in the case of glass, and is probably due to occluded impurities.

When these impurities are driven off by heat, the wire emits no nuclei until it begins to glow. The emission at glowing temperatures is very constant, and does not fatigue. The size and number of the nuclei emitted increase with the temperature.

Similar results were obtained with a wire of ni-chrome, an alloy of nickel and chromium.

Thus in all cases examined where nuclei are driven off from a substance by heat an initial emission takes place at low temperatures which falls off rapidly. The emission of nuclei when the substance glows is practically constant.



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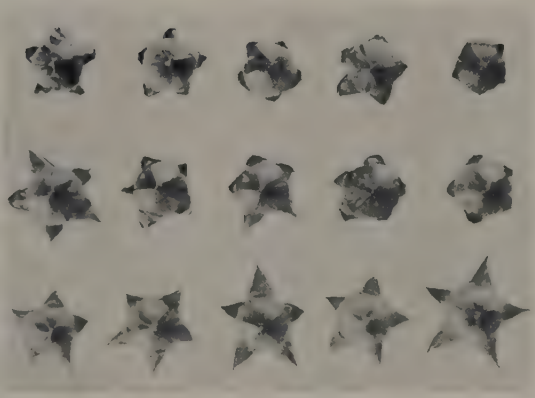

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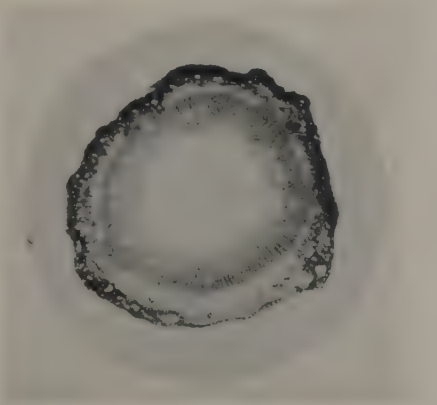

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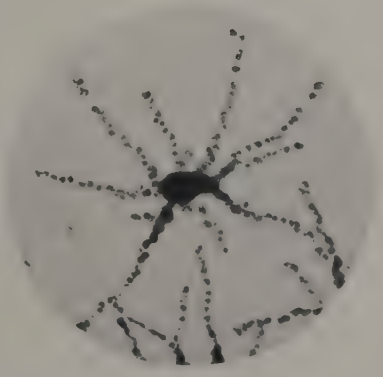

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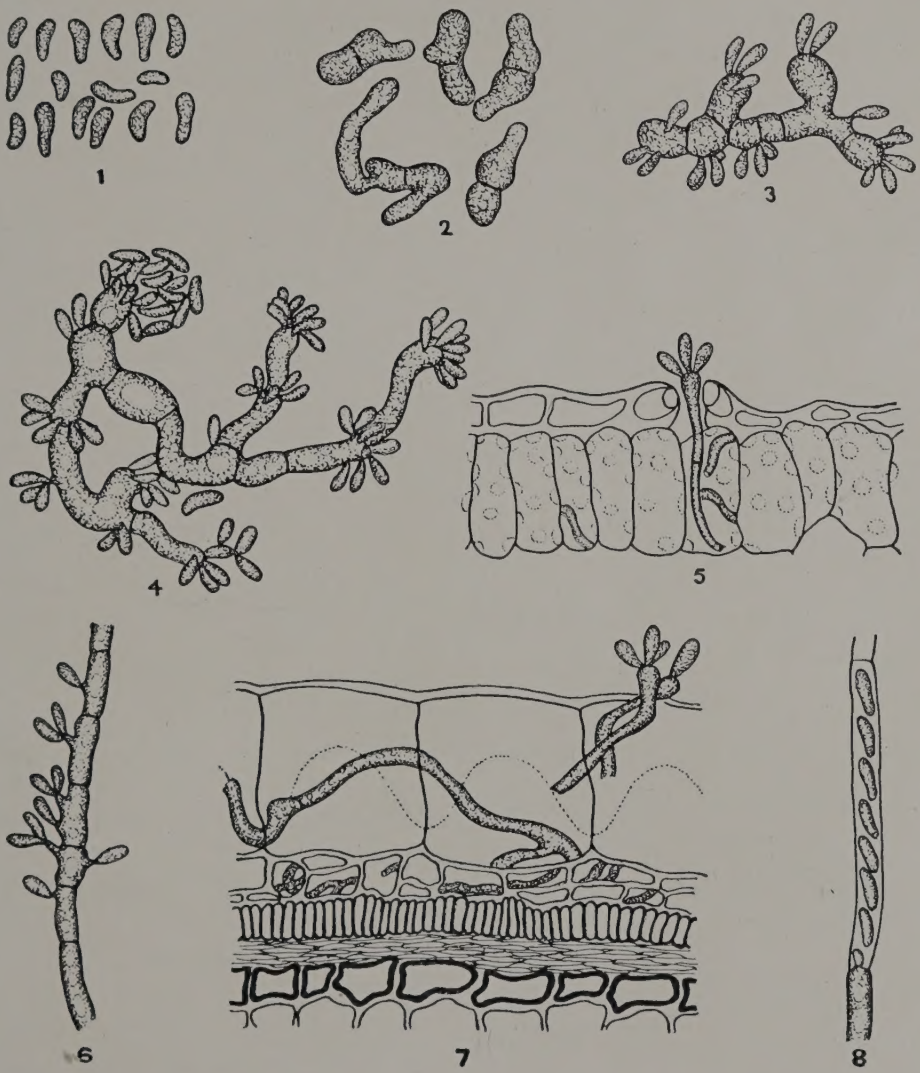
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